Jun 20, 2025

Dear Editor and Reviewers,

Thank you for your insightful comments on this manuscript. We have carefully read each suggestion and given our point-to-point response to all the comments. We believe that the revisions have significantly improved the quality of the manuscript. The changes have been clearly marked in red in the revised version. We sincerely appreciate your efforts in reviewing our work again.

Best Regards,

Tian Feng, PhD

On behalf of all authors

Reply to Anonymous Referee #2:

We thank the reviewer for the careful reading of our manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

General comments

The influences of emission changes, particularly the precursors, on ozone formation have drawn much attention in the science community, but few studies have focused on the photochemical condition related to solar radiation. Since the surface ozone is formed in photochemistry, the change in incident solar radiation is vital for ozone formation. The authors examine the crucial role of changing solar radiation, mostly perturbed by the appearance of clouds, in ozone formation in eastern China. Also, future scenarios of solar radiation and clouds are involved to present a projection of ozone pollution. The authors finally highlight that climate change will pose greater challenges for ozone pollution prevention and control in China. The manuscript is well organized with clear structure, and the language reads fluent. I still have some concerns in the introduction and future projections as shown in the major comments. The authors should address these issues soundly before its publication. Besides that, some technical revisions follow.

Major comments

1. The introduction on the current knowledge of the impacts of solar radiation on ozone formation is insufficient, although the authors have introduced the meteorological factors that affect ozone formation. More information and references are suggested to be included in the Introduction.

Response: The other reviewer also comments that the introduction lacks of information on the influence of LCC, SSRD and CRI on O_3 production, hindering the understanding on the significance of this study. We would like to address both comments together.

In the revised manuscript, we have rewritten the fourth paragraph of the introduction and supplemented the original content, focusing primarily on how variations in cloud cover and solar radiation affect O_3 formation through altering photolysis rates. Unfortunately, we have not found any reference specifically addressing the impact of cloud–radiation interactions on O_3 formation. This is the most significant contribution of our study. In addition to changes in cloud cover and solar radiation themselves, their interactions are also an important factor influencing O_3 concentration. This factor is closely related to climate change and is of increasing importance for future O_3 pollution control as well as related ecological and health studies.

The revised fourth paragraph is as follows: "*However, ground-level O_3 is inherently a photochemical product, and anthropogenic emissions are source drivers that determine its levels, while incident solar radiation acts as a trigger for photochemical reactions, dominating photolysis rates of O_3 production. Currently, there are few studies on the influence of changes in solar radiation on O_3 formation. Early studies reported that clouds have important impacts on tropospheric photochemistry, which increases global mean OH concentration by about 20% (Tie et al., 2003). It was also found that the prediction accuracy of clouds in the model would significantly affect atmospheric chemical composition near the surface layers, leading to an overestimation/underestimation of O_3 concentration (Pour-Biazar et al., 2007). During the Texas Air Quality Study II Radical and Aerosol Measurement Project, the influence of clouds on photolysis rate was evidently greater than that of aerosols (Flynn et al., 2010), and the total reduction in the photolysis rate caused by clouds and aerosols was almost linearly correlated with the reduction in the net O_3 production. These studies all indicates that changes in clouds and solar radiation significantly influence the photolysis conditions, which is of great importance to O_3 formation. In China, the decline in $PM_{2.5}$ concentration is considered one of the reasons for the increase in O_3 levels in recent years due to the weakened aerosol-radiation interactions (Yang et al., 2022). However, there are lack of field campaign evidences similar to those of the USA (Flynn et al., 2010), and only in recent years, fewer studies have qualitatively described the influence of solar radiation on O_3 concentration. For example, enhanced solar radiation during hot and dry weather can increase O_3 production (Mousavinezhad et al., 2021; Xia et al., 2022; Yin et al., 2019; Zhao and Wang, 2017). Some of these studies have also mentioned that cloud cover can alter solar radiation, thereby affecting O_3 formation (Xia et al., 2022; Zhao and Wang, 2017). Nonetheless, these studies are lack of quantitative analysis and systematic mechanism explanations of the contributions of clouds, solar radiation, and their variability to O_3 formation, and none of them further investigate the impact of cloud-radiation interactions (CRI) on O_3 formation. Moreover, with an increasingly persistent impact of climate change, how this factor may affect O_3 concentration remains unclear.*"

The added references have been included in the reference list of the revised manuscript.

Flynn, J., Lefer, B., Rappenglück, B., Leuchner, M., Perna, R., Dibb, J., Ziemba, L., Anderson, C., Stutz, J., Brune, W., Ren, X., Mao, J., Luke, W., Olson, J., Chen, G. and Crawford, J.: Impact of clouds and aerosols on ozone production in Southeast

Texas, Atmos. Environ., 44(33), 4126–4133, 2010.

Pour-Biazar, A., McNider, R., Roselle, S., Suggs, R., Jedlovec, G., Byun, D., Kim, S., Lin, C., Ho, T., Haines, S., Dornblaser, B., Cameron, R.: Correcting photolysis rates on the basis of satellite observed clouds, J. Geophys. Res., 112, D10302, doi:10.1029/2006JD007422, 2007.

Yang, H., Chen, L., Liao, H., Zhu, J., Wang, W. and Li, X.: Impacts of aerosol–photolysis interaction and aerosol–radiation feedback on surface-layer ozone in North China during multi-pollutant air pollution episodes, Atmos. Chem. Phys., 22(6), 4101–4116, doi:10.5194/acp-22-4101-2022, 2022.

2. The comparison between the scenarios in 2021 and 2022 suggests crucial impacts of solar radiation on ozone. This result has a background that the anthropogenic emissions change little for these two years. This is indeed right because the two years are so close that the emissions shall not change much. Yet, for the SSP scenarios in the 21st century, emissions are expected to vary largely during the long term. What will the ozone pollution be considering both the changes in emissions and solar radiation? I suggest the authors to include more discussion on it.

Response: Based on recent emission inventories, both VOCs and NO_x emissions in China have shown a decreasing trend. In this study, by comparison with emissions in the summer of 2021, VOCs and NO_x emissions in the summer of 2022 decreased by 4% and 5%, respectively, leading to a reduction in O_3 concentration by 1.5 μg m^{-3}. According to the current rates of emission reductions, using a simple linear extrapolation, in conjunction with China's carbon neutrality goal, we estimated that VOCs and NO_x emissions will have decreased by 31% and 37%, respectively, relative to 2021 levels by the 2030 carbon peak. By the 2060 carbon neutrality goal, the reductions are projected to reach 80% and 87%, respectively. Clearly, in reality, emission reductions may face challenges and are unlikely to follow a perfectly linear trend, and the O_3 response to precursor reductions is also nonlinear. Nonetheless, here we assume that, in accordance with this idealized scenario, the consequent reduction in O_3 concentrations by 2030 and 2060 are estimated to be reduced by 13.5 μg m^{-3} and 58.5 μg m^{-3}, respectively. Therefore, in the long term, on the decadal scale, the continued emission reductions are expected to effectively control O_3 pollution, leading to significantly lower O_3 concentrations compared to current O_3 levels.

Under three different SSPs, variabilities of clouds and SSRD in summer show a favorable environment for O_3 formation, with an increase rate of SSRD from 0.21 to 0.22 W m^{-2} per year, while interannual variability can reach several tens of W m^{-2}. In this study, interannual difference in SSRD between the summers of 2022 and 2021 is more than 80 W m^{-2}. According to the linear relationship between O_3 and SSRD shown in Figure 2, such differences in SSRD corresponds to a change of 28 μg m^{-3} in daytime O_3 concentration. According to the spatial distribution in Figure 4, the regional mean daytime O_3 change due to meteorological changes (including SSRD) is 9.2 μg m^{-3}. Thus, in the short term, on an interannual scale, the SSRD variability, particularly a sudden increase in SSRD, may partially offset the benefits of emission reductions. Given that coordinated VOCs and NO_x emission reductions are in the early stage, highly favorable photochemical conditions could not only counteract the effects of emission reductions, but may even lead to a rebound in O_3 concentrations.

In the last paragraph in **Section 3.4**, we added discussions on the possible changes in O_3 concentration caused by the changes in emissions and solar radiation, and the text is "*Fortunately, based on recent emission inventories, pollutants in China have shown a decreasing trend. In our study, by comparison with emissions in the summer of 2021, VOCs and NO_x emissions in the summer of 2022 decreased by 4% and 5%, respectively (Jiang et al., 2022; Li et al., 2024), leading to a reduction in O_3 concentration by 1.5 μg m^{-3}. According to these emission reduction rates, we use a simple linear extrapolation method, also in conjunction with China's carbon neutrality goal, to estimate VOCs and NO_x emissions in the future. By the 2030 carbon peak, VOCs and NO_x emissions will have been reduced by approximately 31% and 37%, respectively, relative to 2021 levels. By the 2060 carbon neutrality goal, the reductions are projected to reach 80% and 87%, respectively. Actually, emission reductions may face challenges and unlikely to follow such a perfect pathway, and the response of O_3 concentration to precursor reductions is also nonlinear. We thus assume that, if such an idealized scenario is followed, O_3 concentrations by 2030 and 2060 are estimated to be reduced by 13.5 μg m^{-3} and 58.5 μg m^{-3}, respectively, relative to the levels in 2021. Therefore, in the long term, on the decadal scale, the continued emission reductions are expected to significantly reduce O_3 concentration.*

However, on an interannual scale, the projected SSRD variability can reach several tens of W m^{-2}, which is consistent with this study. Our study shows that interannual *difference in SSRD between the summers of 2022 and 2021 is more than 80 W m^{-2}. Based on the linear relationship between O_3 and SSRD shown in Figure 2, such differences in SSRD corresponds to a change of 28 μg m^{-3} in daytime O_3 concentration. According to the spatial distribution in Figure 4, the regional mean daytime O_3 change due to meteorological changes (including clouds and SSRD) is 9.2 μg m^{-3}. Thus, a sudden increase in SSRD may partially offset the benefits of emission reductions. Given that coordinated VOCs and NO_x emission reductions are in the early stage, the increasing possibility of highly favorable photochemical conditions under climate change could not only counteract the effects of emission reductions, but may even lead to a rebound in O_3 concentrations in the short term.*".

3. We know that the SSP scenarios have large uncertainties. In Figure 6, the SSP scenarios are shown from 2025 to 2099 only. As the first quarter of the 21st century has past, are these SSP scenarios consistent with the variations in real-world Tmax, TCC, and SSRD? The ERA5 reanalysis may help.

Response: We have redrawn Figure 6 to include a comparison analysis between the SSPs projections and the ERA5 reanalysis data for the past 10 years (SSPs projections have been available since 2015). Although there is a noticeable deviation between the projections and observations (reanalysis datasets are generated by assimilating multi-source observation data), the SSPs projections can still provide meaningful information for predicting future ozone pollution trends.

We have added some discussions about the uncertainties of the SSPs scenarios in Lines 456-465: "*The projected climate change under each SSP deviates significantly from the ERA5 reanalysis data, particularly in terms of the interannual variability, which is remarkably larger in reality. This indicates that climate change is highly uncertain. Nevertheless, the projected trend of T_max is generally consistent with the ERA5. The TCC pattern also align well with the SSP2-4.5 projection in recent years, and the SSRD pattern also closely matches the SSP2-4.5 projection. This consistency roughly corresponds with the development pathway in China over the past decade. These comparisons suggest that the projections under different SSPs provide valuable information on understanding future climate change and its implications for O_3 pollution.*".

4. The model validation approach should appear in Data and Method.

Response: We have moved the model validation method from ***Section* Model validation** to ***Section* Data and Methods**, with minor revisions. The revised text is "*To evaluate the model performance, we use three common statistical indices involving mean bias (MB), root mean square error (RMSE), and index of agreement (IOA) (Willmott, 1981).* The formulas are as follows:

$$MB = \frac{1}{N}\sum_{i=1}^{N}(P_i - O_i) \quad (1)$$

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2\right]^{\frac{1}{2}} \quad (2)$$

$$IOA = 1 - \frac{\sum_{i=1}^{N}(P_i - O_i)^2}{\sum_{i=1}^{N}(|P_i - \overline{O}|+|O_i - \overline{O}|)^2} \quad (3)$$

where P_i and O_i represented the simulated and observed variables, respectively. N is the total sample number of the simulation, and $\bar{O}$ denotes the average of the observation. The IOA ranges from 0 to 1. The closer it is to 1, the better the simulation."

Minor comments

5. L45-48: need references, also for L48-49

Response: References have been added.

Lines 48-49 and Lines 50-51 present conclusions from the same reference of Xiao et al. (2022). In the revised manuscript, we have reorganized these two sentences into a single, cohesive statement as follows: "*Another notable aspect is that high O_3 concentration often coincides with high-temperature weather, and their co-occurrence frequency has increased at a faster rate than either alone in recent years (Xiao et al., 2022)*".

Reference

Zhang, Y. and Zheng, J.: Blue Book on Ozone Pollution Prevention and Control in China (2020) [in Chinese], edited by Li, M., Science Press, Beijing, China, 121 pp., ISBN 9787030716644, 2022.

6. L59: the temperature --> air temperature

Response: Revised.

7. L95: in particular, and --> and, in particular,

Response: Revised.

8. L227-235: The ozone variation during 2013-2021 is totally attributed to the emission changes in ozone precursors by the authors. Not any other contributors?

Response: Yes, from 2013 to 2021, the inter-annual variation of ozone concentration was influenced not only by emission changes of precursors, but also by meteorological conditions and $PM_{2.5}$ reductions. The meteorological conditions play an important but not dominant role in ozone trends (Li et al., 2020), and the continued $PM_{2.5}$ reduction weakens the aerosol uptake of hydroperoxyl (HO_2) radicals and enhances ozone production (Li et al., 2018). However, the ozone trend is primarily driven by emission changes (Liu et al., 2023; Liu and Wang, 2020). Our main purpose in citing multiple studies on the impact of emission changes on ozone trends is to demonstrate that the effect of emission changes on ozone trends could potentially be offset by changes in solar radiation conditions in the future (This is one of the main conclusions of our study).

To avoid the misunderstanding that O_3 variation is solely caused by changes in precursor emissions, we have revised and supplemented this section in Lines 263-278: *"Due to the Action Plan on Prevention and Control of Air Pollution since 2013, China's anthropogenic NO_x emissions were substantially reduced (Zhang et al., 2019), whereas VOCs emissions increased slightly during 2013-2017 (Zheng et al., 2018). The disproportionate emission reductions largely contributed to the continuous increase in O_3 concentration from 2013 to 2017 (Jiang et al., 2022; Wang et al., 2022; 2020). Since 2017, as VOCs emissions began to decline (Jiang et al., 2022; Simayi et al., 2022), along with the ongoing reduction in NO_x (Li et al., 2024; Zheng et al., 2018), O_3 concentration began to decline (Lu et al., 2019). In addition to precursor emissions, O_3 trends during this period were also influenced by meteorological conditions and $PM_{2.5}$ reductions. The meteorological conditions play an important but not dominant role in ozone trends (Liu et al., 2023; Li et al., 2020), and the continued $PM_{2.5}$ reduction enhances ozone production due to the weakened aerosol uptake of hydroperoxyl (HO_2) radicals (Li et al., 2019a). Nevertheless, O_3 trends was primarily driven by changes in precursor emissions (Wang et al., 2022a; Liu and Wang, 2020b)."*

In addition, to avoid the redundance with the revised text, we have slightly adjusted the beginning of the following paragraph. "*According to the principle of O_3 formation, it is influenced not only by changes in precursor emissions but also by the solar radiation intensity.*"

New citations have been also added in the reference list in the revised version.

Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q. and Bates, K. H.: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China, vol. 116, pp. 422–427. 2019a.

Li, K., Jacob, D. J., Shen, L., Lu, X., De Smedt, I. and Liao, H.: Increases in surface ozone pollution in China from 2013 to 2019: anthropogenic and meteorological influences, Atmos. Chem. Phys., 20(19), 11423–11433, doi:10.5194/acp-20-11423-2020, 2020.

Liu, Y., Geng, G., Cheng, J., Liu, Y., Xiao, Q., Liu, L., Shi, Q., Tong, D., He, K. and Zhang, Q.: Drivers of Increasing Ozone during the Two Phases of Clean Air Actions in China 2013-2020, Environmental Science & Technology, 57(24), 8954–8964, doi:10.1021/acs.est.3c00054, 2023.

Liu, Y. and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 - Part 2: The effects of emission changes and implications for multi-pollutant control, Atmos. Chem. Phys., 20(1), 6323–6337, doi:10.5194/acp-20-6323-2020, 2020b.

9. L253-254: Revise the citation format

Response: Revised.

10. L289: decimal precision --> decimal fractions

Response: Revised. We have revised "*Additionally, ……without decimal precision*" to "*Additionally, ……, with no decimal fractions*".

11. L293-294: a should be an. I also see this typo in other lines of the text.

Response: We have corrected all similar misuses throughout the text.

12. L305: MB = -0.0 --> MB = 0.0

Response: Corrected. The MB is a negative number, and is approximately 0 after rounding to one decimal place. To indicate the direction of the deviation, a negative sign is included. We have removed the negative sign "-" in the revised version.

13. L365: Noticeably, the correlation between O_3 concentration and SSRD is more significant. Compare with what?

Response: Sorry, here is an error in the previous statement. The correct meaning is that the correlation between O_3 concentration and SSRD is less more significant than the correlations in Figures S6a and S6b.

We have corrected the sentence in Lines 402-406: "… *the correlation between O_3 concentration and SSRD is less more significant than the correlations in Figures S6a and S6b, with a confidence level exceeding 95% (whereas the first two panels show confidence levels exceeding 99.9%). The data are also distributed more dispersedly.*".

14. Figure 1, L759: remove 'map'; L764: the MEGAN --> MEGAN

Response: Removed.