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 #1

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

In this paper, the authors conducted an analysis of the O_3 observation and reanalysis data, employing WRF-Chem to quantify the contribution of CRI to O_3 generation during the 2022 warm season. Furthermore, potential future O_3 pollution risks are assessed based on the CMIP6. The manuscript's structure and the English expression is good. I would like to review the following major comments.

General Comments

1. This study focuses on the effect of CRI on O_3. Nevertheless, a paucity of relevant reviews exists regarding the effects of LCC, SSRD and CRI on O_3 production in the introduction. This deficiency hinders the comprehension of the significance of the work for readers, and consequently, the recommendation is made that relevant content should be incorporated.

Response: The other reviewer also comments that the introduction lacks of information on the influence of solar radiation on O_3 formation. Therefore, we address both comments together. 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 references specifically addressing the impact of cloud–radiation interactions on O_3 formation. This is also 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.*".

In addition, the framework of this study presented in the last paragraph of the Introduction has also been revised accordingly. The revised text is "*Using numerical models, we analyze the causes of high* O_3 *concentration and, in particular, assess the dependence of* O_3 *change on the variabilities of clouds, solar radiation and CRI. Furthermore, we project the potential impacts of these factors on high* O_3 *concentration under climate change.*".

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 authors conducted a comprehensive analysis of various factors leading to the abnormal increase in O_3 concentration in July 2022, and emphasized the importance of CRI in them. It is recommended that the authors undertake a comparative analysis of the contribution of CRI to O_3 change with that of other influencing factors, and incorporate a discussion on the relevant mechanism to enhance the clarity of the analysis.

Response: We have added a new figure (Figure S8 shown below) to specifically illustrate the contributions of different factors to O_3 change, and explained how CRI affects O_3 change by comparing its contributions with those of other factors. Figure S8 is based on Figure 3 and Figure 5c in the main text. The contribution of meteorological changes (orange bar in Figure S8) is calculated as the difference between the orange and green bars in Figure 3, while the contribution of emission changes (blue bar in Figure S8) is the difference between the orange and red bars in Figure 3. The contribution of CRI change (green bar in Figure S8) is the regional mean O_3 change in the YRD shown in Figure 5c.

We have included the related sentences in Lines 434-452: "*Such changes in LCC and SSRD can lead to variations in the CRI, resulting in significantly different impacts on O_3 production. Compared with the summer of 2021, the weaker CRI in the summer of 2022 leads to a widespread and substantial increase in O_3 change over the YRD, with the maximum increase exceeding 9 μg m^{-3} on a local scale (Figure 5c). This implies that a weakened CRI suppresses O_3 formation less effectively, thereby indirectly enhancing O_3 production, with a regional mean O_3 increase of 2.9 μg m^{-3} (Figure S8).*

Based on the above results, contributions of different factors to O_3 increase over the YRD in the summer of 2022 are shown in Figure S8. Changes in meteorological conditions including the reduction in LCC and the increase in SSRD lead to an increase of 9.2 μg m^{-3} in O_3 concentration. Thereinto, the weakened CRI due to the reduced LCC and increased SSRD contributes to 2.9 μg m^{-3}, accounting for 31.5% of the total O_3 increase caused by favorable meteorological conditions. In contrast, anthropogenic VOCs and NO_x emission reductions lead to a decrease of 1.5 μg m^{-3} in O_3 concentration, which is far less than the impact of the changes in photolysis conditions. This indicates that the reduction in LCC, the increase in SSRD, and the weakened CRI are the major drivers of the sudden increase in O_3 concentration over the YRD during the summer of 2022.".

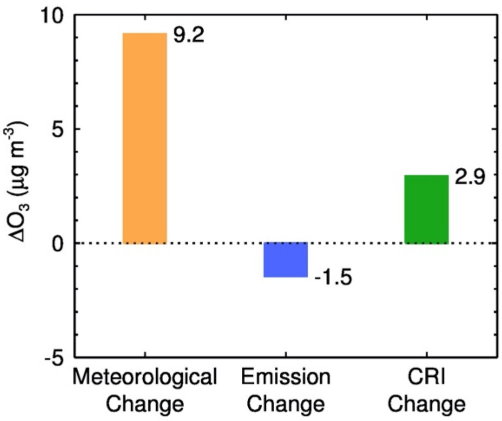

Figure S8 Contributions of interannual variability in various influence factors to ΔO_3. These factors include meteorological conditions, precursor emissions, and CRI. The changes in meteorological conditions also refer specifically to variabilities in LCC and SSRD.

3. As demonstrated in Section 3, the analysis indicates that CRI has been shown to contribute significantly to the abnormal increase in O_3 during the warm season of 2022. As the climate continues to warm in the future, lower LCC and higher SSRD are evident in CMIP6 products; however, the precise contribution of future CRI changes to future O_3 trends remains unclear. It is recommended that the authors incorporate a more explicit discussion.

Response: The major highlight of this study is that we identify the CRI intensity as a new factor to affect change in O_3 concentration, but we have to acknowledge that the precise contribution of future CRI changes to O_3 trends requires more studies for validation. To address the reviewer's comment, we would like to suggest that the interannual differences of summer SSRD and cloud cover under SSPs projections could reach or even exceed those observed between the summers of 2022 and 2021 (in this study, the interannual difference of SSRD more than 80 W m^{-2} and cloud cover difference reaching 0.09). This may indicate that the impact of interannual variation of CRI intensity on O_3 concentration change (ΔO_3) in the future could be no less than what was calculated in this study (ΔO_3 = 2.9 μg m^{-3}).

Related discussions have been added to the final paragraph of **Section 3.4** in Lines 523-530: "*Moreover, less clouds and more SSRD under SSPs will also weaken CRI and consequently aggravate O_3 pollution in the future. Based on SSPs projections, the interannual differences of summer SSRD and cloud cover could reach or even exceed those observed between the summers of 2022 and 2021 (the interannual differences in SSRD and LCC is 82.7 W m^{-2} and 0.09, respectively). It is reasonable to expect that the CRI interannual variability will likely exert an influence on O_3 changes that is no less significant than the calculation presented in this study.*"

Specific Comments

4. Page 2, Line 19-31 It is recommended that numerical descriptions be included to facilitate a more profound comprehension of the impact of CRI on O_3 among scholars.

Response: We have added the information on numerical experiments and rewritten parts of the Abstract, to make readers understanding the highlights more clearly. The revised text in Lines 20-31 is "*Here, we use a regional atmospheric chemistry model, along with 10-year ground-level O_3 measurements, and reanalysis data on low cloud cover (LCC) and surface downward shortwave radiation (SSRD) to investigate the impacts of variations in LCC, SSRD and cloud-radiation interactions (CRI) on O_3 production. We design six numerical experiments, and specifically modify parameters related to cloud radiation effects in the chemistry module to find out the underlying cause for O_3 increase during the warm season of 2022 in the Yangtze River Delta (YRD), China. Results show that O_3 production is strongly modulated by LCC and SSRD. The CRI plays a significant role in regulating O_3 concentration, i.e., reduced LCC, increased SSRD, and a weakened CRI are primarily responsible for the sharp increase in warm-season O_3 concentration observed in 2022 in the YRD, China.*".

5. Sect.2.2, Page 5, Line 174-175 Table S2 is recommended to be placed in the Figures to facilitate the reading process for the reader.

Response: We have moved Table S2 from to the Supplement to the main text and renumbered it as Table 1. Accordingly, the numbering of the other tables has been updated, and the original Table S3 is now Table S2. In addition, the reviewer suggested that CRI should be strengthened in scenario design by numbering scenarios with or without consideration of CRI separately (the last comment). Table 1 shown below has been revised according to the two specific comments.

Table 1 Setup of model experiments.

Experiment	Anthropogenic emission	Meteorology	Cloud-radiation interactions (CRI)
BS Exp._CRI	Emission 2022	Meteorology 2022	Yes
BS Exp._noCRI	Emission 2022	Meteorology 2022	No
CTRL Exp.1_CRI	Emission 2022	Meteorology 2021	Yes
CTRL Exp.1_noCRI	Emission 2022	Meteorology 2021	No
CTRL Exp.2	Emission 2021	Meteorology 2022	Yes
BG Exp.	No	Meteorology 2022	Yes

6. Sect.3.3, Page 10, Line 353-355 In order to enhance the clarity of the data, it is suggested that a greater emphasis be placed on the comparison of LCC and SSRD values.

Response: LCC and SSRD are indeed the most directly meteorological factors affecting O_3 formation, and more accurate model validation is therefore necessary. Unfortunately, LCC and SSRD are not routinely meteorological parameters observed at ground-level weather stations, and are difficult to obtain. As an alternative, we use reanalysis data to evaluate the performance of the model in simulating regional mean LCC and SSRD. To emphasize the comparison of LCC and SSRD, we have added a comparative analysis between observations and simulations in the revised version. We have supplemented the percentage deviations, and discussed how such biases may influence the assessment on the impacts of LCC and SSRD variabilities on O_3 formation.

We have added the sentences in Lines 390-395: "*These comparisons mean that the calculated interannual variability of LCC is approximately 22.2% lower than the observations, while SSRD variability is overestimated by about 1.0%. This may lead to a little underestimation of the impact of LCC and SSRD variabilities on O_3 formation. Generally, the model evidence confirms the observed linkage that an increase (decrease) in LCC and a decrease (increase) in SSRD can suppress (enhance) O_3 production (Figure S6).*".

7. In Table S2, the authors have devised a series of scenarios with the objective of quantifying the influence of different factors on O_3. Given the focus of this paper on the contribution of CRI to O_3 generation, it is suggested that CRI should be strengthened in scenario design, for example by numbering scenarios with or without consideration of CRI separately.

Response: The scenarios with and without the impact of CRI on O_3 formation have been assigned separate labels in Table 1 (Table S2 in the original manuscript). Table 1 has been shown above. The newly description have been added in **Section 2.2 Model and experiments**, and the related text is as follows: "*The BS_Exp. experiments with CRI considered or not are designated as BS_Exp._CRI and BS_Exp._noCRI, respectively, while the CTRL_Exp.1 experiments with and without CRI are designated as CTRL_Exp.1_CRI and CTRL_Exp.1_noCRI. The setup information for all simulation experiments is provided in Table 1.*".