Cloud-radiation interactions amplify ozone pollution in a warming climate

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18   **Abstract**

19   Ozone (O_3) pollution has recently become the most critical air quality issue in China,

20   yet its underlying drivers related to climate change remain poorly understood. Here, we

21   use a regional atmospheric chemistry model, along with 10-year ground-level O_3

22   measurements, and reanalysis data on low cloud cover (LCC) and surface downward

23   shortwave radiation (SSRD) to investigate the impacts of variations in LCC, SSRD and

24   cloud-radiation interactions (CRI) on O_3 production. We design six numerical

25   experiments, and specifically modify parameters related to cloud radiation effects in

26   the chemistry module to find out the underlying cause for O_3 increase during the warm

27   season of 2022 in the Yangtze River Delta (YRD), China. Results show that O_3

28   production is strongly modulated by LCC and SSRD. The CRI plays a significant role

29   in regulating O_3 concentration, i.e., reduced LCC, increased SSRD, and a weakened

30   CRI are primarily responsible for the sharp increase in warm-season O_3 concentration

31   observed in 2022 in the YRD, China. Moreover, climate warming is likely to exacerbate

32   future O_3 pollution via weakening CRI due to fewer clouds and more SSRD. To

33   mitigate O_3 pollution, we thus propose implementing more stringent emission reduction

34   measures on O_3 precursors, along with proactive strategies to address climate change.

35

36   **Short Summary**

37   This study investigated how cloud-radiation interactions influence ozone formation in

38   a warming climate. Using measurements, reanalysis data and models, we found that

39   cloud-radiation interactions can worsen O_3 pollution and climate warming will amplify

40   the influence. We highlight that climate change will pose greater challenges for China's

41   O_3 pollution prevention and control, and actions such as reducing O_3 precursors

42   emissions and mitigating climate change are urgently needed.

43

44   **Keywords**: Ozone, cloud-radiation interactions, climate change, emission reduction

45

1 Introduction

Over the past decade, high concentrations of ground-level ozone (O_3) have increasingly been a major air pollution issue in China. These O_3 pollution events are characterized by extensive spatial coverage and prolonged duration during the warm season, i.e., from September 22 to 29, 2019, a severe O_3 pollution event in eastern China covered an area of approximately 3.2 million square kilometers (Zhang and Zheng, 2022). 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), posing serious risks to human health, climate change, and food security.

Ground-level observations show that each 1°C increase results in an 8 - 10 μg m^{-3} rise in O_3 concentrations during heatwaves in eastern China, when air temperature varies between 28°C and 38°C (Pu et al., 2017; Wang et al., 2023). This is largely attributed to O_3 sensitivity to the precursors. In the VOC-limited regime, an increase in air temperature can enhance biogenic VOCs emissions, providing more O_3 precursors (Liu and Wang, 2020a). However, the response of O_3 concentration to air temperature is nonlinear. As the temperature further increases and exceeds 38.5°C, chemical and biophysical feedbacks of vegetation are suppressed, and consequently, biogenic emissions and related O_3 formation are reduced (Meehl et al., 2018; Pu et al., 2017; Steiner et al., 2010). Thus, extreme high temperature cannot fully explain high O_3 concentration. What exactly causes the highest daytime O_3 concentration in the hottest summer?

Several recent review studies have identified multiple factors to explain ground-level O_3 formation (Fu et al., 2019; Jiang et al., 2022; Lu et al., 2019a; Wang et al., 2022a), including precursor emissions and their proportion (Mousavinezhad et al., 2021; Wang et al., 2019b; Xue et al., 2014; Zeng et al., 2018; Zheng et al., 2023), climate patterns (Creilson et al., 2005; Gao et al., 2023; Hong et al., 2019; Shen and Mickley, 2017; Xu et al., 2017), synoptic-scale circulation systems (Dong et al., 2020; Ji et al., 2024; Jiang et al., 2021; Li et al., 2018; Mao et al., 2020; Shu et al., 2016; Yin et al., 2019; Zhao et al., 2010; Zhao and Wang, 2017; Zheng et al., 2023; Zhou et al., 2013), meteorological parameters such as temperature (Lu et al., 2019b; Mousavinezhad et al., 2021; Pu et al., 2017; Wang et al., 2023; Zheng et al., 2023), humidity (Mousavinezhad et al., 2021; Pu et al., 2017; Zhao and Wang, 2017; Zheng et al., 2023), wind (Mao et al., 2020; Pu et al., 2017; Zhao and Wang, 2017), and boundary layer height (Mousavinezhad et al., 2021; Zheng et al., 2023), as well as stratosphere-troposphere exchange (Lu et al., 2019a; 2019b; Verstraeten et al., 2015).

85 However, ground-level O_3 is inherently a photochemical product, and anthropogenic
86 emissions are source drivers that determine its levels, while incident solar radiation acts
87 as a trigger for photochemical reactions, dominating photolysis rates of O_3 production.
88 Currently, there are few studies on the influence of changes in solar radiation on O_3
89 formation. Early studies reported that clouds have important impacts on tropospheric
90 photochemistry, which increases global mean OH concentration by about 20% (Tie et
91 al., 2003). It was also found that the prediction accuracy of clouds in the model would
92 significantly affect atmospheric chemical composition near the surface layers, leading
93 to an overestimation/underestimation of O_3 concentration (Pour-Biazar et al., 2007).
94 During the Texas Air Quality Study II Radical and Aerosol Measurement Project, the
95 influence of clouds on photolysis rate was evidently greater than that of aerosols (Flynn
96 et al., 2010), and the total reduction in the photolysis rate caused by clouds and aerosols
97 was almost linearly correlated with the reduction in the net O_3 production. These studies
98 all indicates that changes in clouds and solar radiation significantly influence the
99 photolysis conditions, which is of great importance to O_3 formation. In China, the
100 decline in $PM_{2.5}$ concentration is considered one of the reasons for the increase in O_3
101 levels in recent years due to the weakened aerosol-radiation interactions (Yang et al.,
102 2022). However, there are lack of field campaign evidences similar to those of the USA
103 (Flynn et al., 2010), and only in recent years, fewer studies have qualitatively described
104 the influence of solar radiation on O_3 concentration. For example, enhanced solar
105 radiation during hot and dry weather can increase O_3 production (Mousavinezhad et al.,
106 2021; Xia et al., 2022; Yin et al., 2019; Zhao and Wang, 2017). Some of these studies
107 have also mentioned that cloud cover can alter solar radiation, thereby affecting O_3
108 formation (Xia et al., 2022; Zhao and Wang, 2017). Nonetheless, these studies are lack
109 of quantitative analysis and systematic mechanism explanations of the contributions of
110 clouds, solar radiation, and their variability to O_3 formation, and none of them further
111 investigate the impact of cloud-radiation interactions (CRI) on O_3 formation. Moreover,
112 with an increasingly persistent impact of climate change, how this factor may affect O_3
113 concentration remains unclear.

114 In this study, we establish correlations between daytime O_3 concentration and
115 downward solar radiation as well as low clouds, based on measurements and reanalysis
116 data during the past decade. Using numerical models, we analyze the causes of high O_3
117 concentration and, in particular, assess the dependence of O_3 change on the variabilities
118 of clouds, solar radiation and CRI. Furthermore, we project the potential impacts of
119 these factors on high O_3 concentration under climate change.

120
121 **2   Data and Methods**
122 **2.1 Measurements and reanalysis data**
123 We collect *in-situ* measurements on hourly mass concentrations of gaseous pollutants
124 in the Yangtze River Delta (YRD), China during the warm season of the past decade

(2014-2024). The gaseous pollutants include O_3, CO and NO_2, which are measured by Model 49i UV Photometric Ozone (O_3) Analyzer, Model 48i Gas Filter Correlation Carbon Monoxide (CO) Analyzer, and Model 42i Chemiluminescence NO-NO_2-NO_x Analyzer, respectively. These analyzers are equipped with built-in calibration systems that accurately linearize the instrument outputs. The missing data have been eliminated. The flat YRD region, located in eastern China, is one of the largest urban agglomerations in the world, consisting of three provinces (Jiangsu, Zhejiang, and Anhui) and one municipality (Shanghai) (Figure 1a). This region is densely populated, with highly developed economies and transportation networks, and concurrently, anthropogenic emissions of O_3 precursors, including nitrogen oxides (NO_x) and volatile organic compounds (VOCs) are significantly higher than those in other regions of China (Figures 1b and 1c). Moreover, the region has abundant vegetation, resulting in a moderate level of biogenic VOCs emission in China (Figure 1d). Thus, this region is one of China's hotspots for O_3 pollution. The warm season in mid-latitude regions of Northern Hemisphere often refers to the April -September period. In these six consecutive months within a single calendar year, the highest mean O_3 concentration is observed, defined as the warm-season O_3. The YRD belongs to the mid-latitude region (Figure 1a), and is facing an environment issue of high O_3 concentration during the warm season.

Meteorological reanalysis data used here consist of surface downward shortwave radiation (SSRD) and low cloud cover (LCC) from the European Centre for Medium-Range Weather Forecasts ERA5, with an hourly resolution and a 0.25° × 0.25° spatial resolution. SSRD and LCC data are selected from 07:00 to 18:00 Beijing Time (BJT) due to O_3 photochemical formation occurred during the daytime. Hourly observations on 2-m temperature (T2m), relative humidity (RH), wind speed (WS) and direction (WD) observed at four weather stations are from the National Oceanic and Atmospheric Administration, available on the website of https://www.ncei.noaa.gov/maps/hourly/.

2.2 Model and experiments

We use a state-of-the-art regional Weather Research and Forecasting Model online coupled with chemistry (WRF-Chem model) to investigate the causes of high O_3 concentration during the warm season of 2022. The WRF-Chem model is a regional atmospheric chemistry transport model that can assess how the physical and chemical processes including transport, vertical mixing, aerosol-cloud interactions, cloud-radiation interactions, emissions, and gas-to-particle conversion affects air quality. The detailed model information refers to Grell et al. (2005), and model configurations used in this study are as follows. The physical mechanisms include the Goddard longwave and shortwave radiation schemes (Dudhia, 1989), the WSM 6-class graupel microphysics scheme (Hong and Lim, 2006), the Mellor-Yamada-Janji (MYJ)

165   planetary boundary layer scheme (Janjić, 2002), the unified Noah land-surface model
166   (Chen and Dudhia, 2001) and Monin-Obukhov surface layer scheme (Janjić, 2002).
167   The chemical mechanisms include a new flexible gas-phase chemical module and the
168   Community Multiscale Air Quality (CMAQ, version 4.6) aerosol module developed by
169   the United States Environmental Protection Agency (U.S. EPA) (Binkowski, 2003),
170   gas-phase reactions of volatile organic compounds (VOCs) and nitrogen oxide (NO_x)
171   by the SAPRC-99 (Statewide Air Pollution Research Center, version 1999), and a non-
172   traditional volatility basis-set (VBS) approach to calculate secondary organic aerosol
173   (SOA) formation (Li et al., 2011b). In addition, HONO production by NO_2
174   heterogeneous reaction is added to improve HO_x ($OH+HO_2$), NO_x, O_3, and SOA
175   simulations (Li et al., 2010). Inorganic aerosols use the ISORROPIA mechanism
176   (version 1.7) (Nenes et al., 1998), in which a SO_2 heterogeneous reaction to sulfate
177   formation on aerosol surfaces is considered (Li et al., 2017a). A fast Tropospheric
178   Ultraviolet and Visible (FTUV) radiation transfer model is used to calculate the
179   photolysis rates (Tie et al., 2003), which can also calculate the impacts of aerosols and
180   clouds on the photochemistry processes (Li et al., 2011a). The wet deposition uses the
181   method in CMAQ (Byun and Ching, 1999) and the dry deposition follows Wesely
182   (1989). Anthropogenic emission inventory uses the Multi-resolution Emission
183   Inventory for China (MEIC) developed by the Tsinghua University (Li et al., 2017b),
184   consisting of industrial, power, transportation, agricultural, and residential sources. The
185   biogenic emissions are calculated by the Model of Emissions of Gases and Aerosol
186   from Nature (MEGAN) (Guenther et al., 2006). The model horizontal resolution is 6
187   km, with 200 grids in the longitude and 200 grids in the latitude. There are 35 vertical
188   sigma levels, with intervals ranging from 50 m near the surface to 500 m at 2.5 km
189   above the ground level, and more than 1 km above 14 km. Initial and boundary
190   meteorological fields in the model are driven by 6-hour 1° × 1°Final Analyses data from
191   National Centers for Environmental Prediction (NCEP FNL). Chemical initial and
192   boundary fields are from a Community Atmosphere Model with chemistry (CAM-
193   Chem) 6-hour output. The spin-up time of the model is 2 days. A brief introduction on
194   the schemes used in this study is shown in Table S1.
195

196   We perform four groups of model experiments, with a total of six simulations (Table
197   1). The baseline experiment (BS_Exp.) reflects the real situation of high O_3
198   concentration. The BS_Exp. uses real emissions and meteorological conditions in July,
199   2022. The rationality for selecting July as the representative month of the warm season
200   is as follows. Firstly, July is typically the most representative month for the warm
201   season in Northern Hemisphere. Moreover, the observed interannual variation in
202   daytime O_3 concentration in July is fully consistent with the interannual variation in
203   warm-season mean O_3 concentration in recent years (Figure S1 and Figure 2a). Most
204   importantly, daytime O_3 concentration in July 2022 is the highest in recent years,

205 significantly higher than the lowest in July 2021, and another identical feature is that
206 daytime O_3 concentration in July 2024 is the second highest. The BS_Exp. is also used
207 to validate the model performance by comparing with the measurements. Another two
208 groups of control experiments (CTRL_Exp.) are used to assess the impacts of
209 interannual variability of meteorology and emission change on O_3 formation. The first
210 group of control experiments selects the year of 2021 with the lowest daytime O_3
211 concentration in recent 5 years, using the same emissions as the BS_Exp. but different
212 meteorological conditions (defined as CTRL_Exp.1). Differences between the
213 CTRL_Exp.1 and BS_Exp. can illustrate the impacts of interannual variability of
214 meteorological conditions on O_3 concentration. The second group uses the same
215 meteorological conditions as the BS_Exp. but different emissions (CTRL_Exp.2).
216 Emission changes in CTRL_Exp.2 are based on the emissions in 2021, and the
217 difference between CTRL_Exp.2 and the BS_Exp. can explain the impact of emission
218 changes of precursors on O_3 formation. In addition, we perform a background
219 experiment (BG_Exp.) with zero anthropogenic emissions to calculate the background
220 O_3 concentration.

221

222 Based upon the BS_Exp. and CTRL_Exp.1, we particularly examine the contribution
223 of CRI intensity to O_3 formation *via* considering and not considering the impact of CRI
224 on atmospheric photochemistry. The BS_Exp. experiments with CRI considered or not
225 are designated as BS_Exp._CRI and BS_Exp._noCRI, respectively, while the
226 CTRL_Exp.1 experiments with and without CRI are designated as CTRL_Exp.1_CRI
227 and CTRL_Exp.1_noCRI. The setup information for all simulation experiments is
228 provided in Table 1. The impacts of clouds on solar radiation are calculated by adjusting
229 three key parameters in the chemical module related to cloud radiative effect: cloud
230 optical depth, single scattering albedo, and asymmetry factor. This approach confines
231 the CRI impact within photochemical reactions, only altering the photolysis rates of
232 photochemical substances directly associated with O_3 formation. Additionally, it avoids
233 the original meteorological fields in the physical module being perturbed by the CRI,
234 which would otherwise complicate the study.

235

236 To evaluate the model performance, we use three common statistical indices involving
237 mean bias (MB), root mean square error (RMSE), and index of agreement (IOA)
238 (Willmott, 1981). The formulas are as follows:

239 $$\text{MB} = \frac{1}{N}\sum_{i=1}^{N}(P_i - O_i) \tag{1}$$

240 $$\text{RMSE} = \left[\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2\right]^{\frac{1}{2}} \tag{2}$$

241 $$\text{IOA} = 1 - \frac{\sum_{i=1}^{N}(P_i - O_i)^2}{\sum_{i=1}^{N}(|P_i - \overline{O}| + |O_i - \overline{O}|)^2} \tag{3}$$

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.

2.3 Climate Scenarios

Using 41-model results from Coupled Model Intercomparison Project Phase 6 (CMIP6) (Table S2), we analyze the long-term trends of monthly surface downwelling shortwave radiation (SSRD), total cloud cover percentage (TCC), and daily maximum air temperature (T_max) in July during 2025-2099 under three Shared Socio-economic Pathways (SSPs). These three SSPs narrate the Green Road with a sustainable development paradigm (SSP1-1.9), middle-of-the road along a historical development pattern (SSP2-4.5), and a highway road with a fossil-fueled development pattern (SSP5-8.5) that represent high, moderate, and low climate mitigation pathways, respectively (Riahi et al., 2017). Finally, we project the potential influence of the solar radiation on the occurrence of high O_3 concentration under these climate scenarios.

3 Results and discussion

3.1 Observed linkage between O_3, incident solar radiation, and low clouds

Over the past decade, the warm-season mean daytime O_3 concentration (hereafter O_3 concentration) in the YRD has shown a distinct rising-falling pattern before 2021, with a turning point in 2017 (Figure 2a). From 2013 to 2017, O_3 concentration increased by 5.9 μg m^{-3} per year, while decreased by 2.5 μg m^{-3} per year during 2017-2021. 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., 2022a; Liu and Wang, 2020b; Wang et al., 2019a). 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; Zhang et al., 2019), O_3 concentration began to decline (Lu et al., 2019b). 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). Unexpectedly, during the warm season of 2022, O_3 concentration suddenly increased to the highest, even surpassing the turning point of 2017 by 3.4 μg m^{-3}. Subsequently, O_3 concentrations dropped during the warm seasons of 2023-2024

281   compared to the same period of 2022, but still remained at relatively high levels. This

282   is seemingly paradoxical to emission reductions in O_3 precursors mentioned above.

283

284   According to the principle of O_3 formation, it is influenced not only by changes in

285   precursor emissions but also by the solar radiation intensity. Observational evidence

286   reveals that interannual variability of warm-season downward solar radiation is highly

287   consistent with the interannual variation of O_3 concentration in the YRD during the past

288   decade. A significant positive correlation between them (*r = 0.85, p < 0.001*, Figure

289   2b) suggests that O_3 concentration indeed strongly depends on the SSRD intensity. Low

290   clouds with small and compact liquid droplets can significantly reflect the solar

291   radiation by their considerable optical thickness (Kang et al., 2020), thereby

292   diminishing photolysis rate and the loadings of tropospheric oxidants (Tie et al., 2003;

293   2019). We examined the relationship between daytime LCC and O_3 concentration, and

294   found that O_3 concentration is more significantly negative with LCC (*r = -0.90, p <*

295   *0.001*, Figure 2c). This suggests that low clouds are of great importance to O_3

296   concentration. Liu and Wang (2020a) suggested that the reduction of cloud cover plays

297   a dominant role in increasing daily maximum 8-hour (MDA8) O_3 concentration in

298   China during 2014-2017 summer. Similarly, daytime LCC in the warm season of 2022

299   dropped to the lowest (LCC = 0.2) during the past decade, with a 23.6% reduction

300   relative to the multi-year mean, while SSRD was significantly more than the multi-year

301   mean by 28.9 W m^{-2} (Figure S2). Thus, solar radiation is vital to O_3 formation, by which

302   an increase (decrease) in LCC intensifies (weakens) the reflection of solar radiation,

303   and decreases (increases) SSRD, inconducive (conducive) to O_3 formation. The

304   favorable solar radiation is likely crucial to the sudden increase in O_3 concentration

305   during the warm season of 2022, though O_3 precursors from anthropogenic emissions

306   have been slashed.

307

308   **3.2 Model validation**

309

310   Observations at weather stations in four provinces and municipality are used to validate

311   the model performance on meteorological fields. Results show that the model well

312   captures spatiotemporal variability of meteorological parameters (Figure S3). For

313   example, the simulated T2m is in good agreement with the observation, with the IOAs

314   in the range of 0.88-0.92. The MBs are within 0.7°C, and the RMSEs are around 2.0°C.

315   Followed by the RH, with IOAs of 0.83 to 0.89. Most importantly, the model

316   successfully captures the WD shift, with the IOAs of 0.79 to 0.89, which is crucial for

317   accurately simulating atmospheric transport and re-distribution of the spatiotemporal

318   variations in pollutants. There are also some biases between the simulations and

319   observations. The model generally overestimates the WS, with the IOAs between 0.60

320   and 0.73, lower than those of the three parameters mentioned above. These

321 discrepancies are partly due to the systematical bias of the WRF-Chem model, which
322 often overestimates the ground-level WS. Additionally, the observation data on WS are
323 recorded only as integers, with no decimal fractions. This lack of precision in
324 observations reduces the temporal variability of the WS, compared to the simulations,
325 thereby resulting in a lower IOA.

326

327 For pollutants, the model also well reproduces temporal variation of O_3, with an MB of
328 2.4 μg m^{-3} and an RMSE of 17.7 μg m^{-3} (Figure S4a). This means the simulation is
329 approximately 2.8% higher than the observation, with an accuracy of 79.3%. The IOA
330 between the simulated O_3 hourly variation and the observation exceeds 0.90 (IOA =
331 0.94), implying for a better performance of the model on chemical reactions. The
332 simulated NO_2 concentration is also in a good agreement with the observation (IOA =
333 0.83), with an MB of 0.9 μg m^{-3} and an RMSE of 5.3 μg m^{-3} (Figure S4b). Inevitably
334 there are also some discrepancies between the simulation and the observation, i.e, the
335 amplitude of the simulated CO concentration is remarkably greater than the observed
336 (Figure S4c). This is largely related to the emission inventory that fails to depict an
337 accurate diurnal cycle of CO emission. The IOA between the simulated and observed
338 CO concentrations is thus relatively lower (IOA = 0.63). However, the simulated mean
339 CO concentration is extremely close to the observation (MB = 0.0 μg m^{-3}), suggesting
340 that the model accurately captures the variability of atmospheric transport. Generally,
341 the model well reproduces temporal variations in meteorological fields, O_3 and related
342 gaseous pollutants (Figures S3-S4), providing sufficient evidence on the rationality of
343 the model.

344

345 **3.3 Modelling evidence on O_3 increase**
346 To verify this hypothesis, we separately distinguished contributions of background
347 fields, anthropogenic emissions and their changes, as well as changes in meteorological
348 fields to O_3 concentration. Meteorological and chemical lateral boundary inputs, and
349 biogenic emissions approximately produce 57.7 μg m^{-3} of O_3 concentration in the YRD
350 in the warm season of 2022, accounting for 55.1% of O_3 concentration (Figure 3).
351 Another study also found that background inputs contributes 39 to 58 μg m^{-3} to
352 summertime MDA8 O_3 concentration in this region (Li et al., 2019b). These results
353 reveal a relatively high level of background O_3 concentration in the YRD, which
354 provides a favorable environmental basis for the occurrence of O_3 pollution. When
355 anthropogenic emissions are included, O_3 concentration increases by 47.1 μg m^{-3},
356 suggesting that human emissions remain a key contributor to O_3 formation.

357

358 We further investigated the impact of changes in anthropogenic emissions on O_3
359 concentration. Based upon the interannual variations in anthropogenic emissions, NO_x
360 and VOCs emissions in the summer of 2022 are approximately reduced by 5% and 4%,

361 respectively, compared to the summer of 2021 (Jiang et al., 2022; Li et al., 2024).
362 Consequently, O_3 concentration decreased by 1.5 μg m^{-3} (Figure 3), meaning that
363 current emission reductions definitely lead to a decline in O_3 concentration. As
364 summertime O_3 sensitivity changes from a VOC-limited regime to a transitional regime
365 in the YRD (Wang et al., 2019a; 2022b; Yin et al., 2019), simultaneous reductions in
366 VOCs and NO_x emissions have become an effective way to reduce O_3 concentration.
367 Nonetheless, the O_3 drop through emission cuts is not as significant as expected.
368 Therefore, the changes in anthropogenic emissions are not responsible for the increase
369 in O_3 concentration in the warm season of 2022, and more stringent measures on
370 emission reductions are needed to achieve a desired O_3 decline.
371
372 Besides the impacts of human emissions, we examined the influence of meteorology
373 change, because the change is of great significance to the trend of O_3 concentration,
374 even exceeding the impact of changes in anthropogenic emissions (Liu and Wang,
375 2020a). As a result, differences in meteorological fields alone lead to a 9.2 μg m^{-3}
376 increase of O_3 concentration in July 2022 relative to the same period of 2021 (Figure
377 3). This is roughly consistent with Ji et al. (2024), who suggested adjustments of
378 meteorological fields lead to an increase in O_3 concentration by 13.0 μg m^{-3} in coastal
379 cities of the YRD in July 2022 compared to 2015-2021. Thus, meteorological
380 conditions in the warm season of 2022 are more favorable for O_3 formation in the YRD.
381 Noticeably, the negative effects of interannual variability of meteorological conditions
382 on O_3 concentration have greatly exceeded the positive effects of precursor emission
383 reductions.
384
385 Furthermore, we specifically assessed the impacts of shortwave solar radiation, low
386 clouds, and CRI on O_3 concentration, because the solar radiation is the direct
387 meteorological factor for O_3 formation. The model also well reproduces the interannual
388 variability of LCC and SSRD, i.e., the calculated changes in LCC and SSRD are 0.07
389 and 83.5 W m^{-2}, respectively, close to the observed 0.09 and 82.7 W m^{-2} (Figure S5).
390 These comparisons mean that the calculated interannual variability of LCC is
391 approximately 22.2% lower than the observations, while SSRD variability is
392 overestimated by about 1.0%. This may lead to a little underestimation of the impact of
393 LCC and SSRD variabilities on O_3 formation. Generally, the model evidence confirms
394 the observed linkage that an increase (decrease) in LCC and a decrease (increase) in
395 SSRD can suppress (enhance) O_3 production (Figure S6). As LCC increase, the SSRD
396 reduces significantly at a rate of more than 40 W m^{-2} per 0.1 increase in LCC (Figure
397 S6a). In particular, the SSRD decreases more rapidly in the early stage when low clouds
398 appear. As a result, the photolysis rate rapidly drops and O_3 production significantly
399 slows down. As LCC further increases, daily mean SSRD falls below 400 W m^{-2},
400 resulting in a noticeable slowdown in photolysis rates, falling to less than 4.0×10^{-3} s^{-1}

401 (Figure S6b). Consequently, the rate of O_3 production slows, and O_3 concentrations are
402 not as high as that occurred in the early stage (Figure S6c). Noticeably, the correlation
403 between O_3 concentration and SSRD is less more significant than the correlations in
404 Figures S6a and S6b, with a confidence level exceeding 95% (whereas the first two
405 panels show confidence levels exceeding 99.9%). The data are also distributed more
406 dispersedly. This is largely due to solar radiation is one key factor influencing O_3
407 production. Precursors emissions and their proportion are the other key factor. In
408 addition, O_3 concentration is also affected by atmospheric transport, deposition, and
409 stratosphere-troposphere exchange mentioned in *Section Introduction*.
410
411 Unfortunately, current models are unable to fully isolate the individual contribution of
412 variability in LCC and SSRD to O_3 production. As a compromise, we managed to
413 examine the response of O_3 concentration to the CRI. In July 2022, clear-sky weather
414 dominates in the YRD, with monthly mean daytime LCC noticeably lower than that in
415 2021 (Figures 4a and 4b). The regional average daytime LCC and SSRD are 0.04 and
416 583.20 W m^{-2}, respectively (Figures 4a and 4c). Compared to July 2021 (Figures 4b
417 and 4d), LCC decreases by 63.6%, while SSRD increases by 16.7%. This clear and
418 cloudless weather favors O_3 formation. Consequently, the magnitude and spatial
419 coverage of high O_3 concentration are significantly larger (Figures 4e and 4f). Less
420 LCC in July 2022 reflects less incident solar radiation, resulting in less attenuation to
421 incident solar radiation and more solar radiation reaching the surface. This minimal
422 impact of clouds on incident solar radiation is defined as a weak CRI. By comparison,
423 more LCC in July 2021 enhances the reflection of incident solar radiation, and
424 consequently, less incident solar radiation reaches the surface, leading to a strong CRI.
425 Whether the CRI is strong or weak, it reduces SSRD and decelerates the photolysis rate,
426 thereby suppressing ground-level O_3 production (Figures 5a, 5b, and S6). The stronger
427 (weaker) the CRI, the more (less) the O_3 reduction. The change in O_3 concentration
428 (ΔO_3) is highly sensitive to the LCC when low clouds are fewer (Figure S6). A little
429 increase in LCC can cause a sharp decline in O_3 production, resulting in a significant
430 reduction in O_3 concentration. For example, when LCC is less than 0.3, an increase of
431 0.1 in LCC approximately leads to a reduction of 3.5 µg m^{-3} in ΔO_3. When LCC is more
432 than 0.3, the photolysis rates decrease to a lower level (Figures S6a and S6b), and ΔO_3
433 drops not as rapidly as that in the initial stage of clouds occurrence, with a decline rate
434 of 2.4 µg m^{-3} for an additional 0.1 increase in LCC (Figure S7). Such changes in LCC
435 and SSRD can lead to variations in the CRI, resulting in significantly different impacts
436 on O_3 production. Compared with the summer of 2021, the weaker CRI in the summer
437 of 2022 leads to a widespread and substantial increase in O_3 change over the YRD, with
438 the maximum increase exceeding 9 µg m^{-3} on a local scale (Figure 5c). This implies
439 that a weakened CRI suppresses O_3 formation less effectively, thereby indirectly
440 enhancing O_3 production, with a regional mean O_3 increase of 2.9 µg m^{-3} (Figure S8).

441

442   Based on the above results, contributions of different factors to O_3 increase over the
443   YRD in the summer of 2022 are shown in Figure S8. Changes in meteorological
444   conditions including the reduction in LCC and the increase in SSRD lead to an increase
445   of 9.2 $\mu g\,m^{-3}$ in O_3 concentration. Thereinto, the weakened CRI due to the reduced LCC
446   and increased SSRD contributes to 2.9 $\mu g\,m^{-3}$, accounting for 31.5% of the total O_3
447   increase caused by favorable meteorological conditions. In contrast, anthropogenic
448   VOCs and NO_x emission reductions lead to a decrease of 1.5 $\mu g\,m^{-3}$ in O_3 concentration,
449   which is far less than the impact of the changes in photolysis conditions. This indicates
450   that the reduction in LCC, the increase in SSRD, and the weakened CRI are the major
451   drivers of the sudden increase in O_3 concentration over the YRD during the summer of
452   2022.

453

454   **3.4 O_3 pollution potential under global warming**
455   We used CMIP6 products to analyze the long-term trends of T_{max}, SSRD, and TCC
456   under SSP5-8.5, SSP2-4.5 and SSP1-1.9 (Figure 6). The projected climate change
457   under each SSP deviates significantly from the ERA5 reanalysis data, particularly in
458   terms of the interannual variability, which is remarkably larger in reality. This indicates
459   that climate change is highly uncertain. Nevertheless, the projected trend of T_max is
460   generally consistent with the ERA5. The TCC pattern also align well with the SSP2-
461   4.5 projection in recent years, and the SSRD pattern also closely matches the SSP2-4.5
462   projection. This consistency roughly corresponds with the development pathway in
463   China over the past decade. These comparisons suggest that the projections under
464   different SSPs provide valuable information on understanding future climate change
465   and its implications for O_3 pollution.

466

467   Ensemble mean T_{max} will continue to rise during the 21st century under any SSPs,
468   whether the extreme or mean T_{max} (Figures 6a and 6b). Noticeably, until the end of the
469   21st century, T_{max} extreme no longer increases significantly and exhibits a fluctuation
470   pattern under SSP1-1.9. However, under the two alternative scenarios, the T_{max} will
471   continuously increase more significantly, with an annual mean rate of 0.3°C per decade
472   (Figures 6a and 6b). While under SSP5-8.5, the temperature will follow a linear
473   increase trend at a faster rate that is more than twice as that under SSP2-4.5. Although
474   there are some differences in the warming rates by models and scenarios, the warming
475   trend is highly consistent.

476

477   Observational evidence shows that climate warming has increased the frequency of
478   high temperatures and O_3 extremes (Wang et al., 2023). Consequently, the frequency
479   of extreme high-temperature events coinciding with high O_3 concentrations, as
480   observed in 2022, may also increase (Hong et al., 2019; Xiao et al., 2022). Heatwaves

481 are often accompanied by adiabatic subsidence, fewer clouds, and stronger solar
482 radiation. Cloud cover shows no significant trend under SSP1-1.9, whereas it decreases
483 significantly under SSP2-4.5 and SSP5-8.5, with a faster decline rate under SSP5-8.5
484 (Figure 6c). Concurrently, SSRD exhibits a significantly increasing trend under three
485 SSPs (Figure 6d). Though in the second half of the 21st century, SSRD fluctuates within
486 a smaller magnitude under SSP1-1.9, it is still higher than that in the first half of the
487 21st century. Under SSP2-4.5 and SSP5-8.5, SSRD increases more rapidly at almost the
488 same rate (Figure 6d).
489
490 There are some differences in the trends of radiation factors related to O_3 formation
491 under different scenarios inevitably. For example, the phases of SSRD and clouds under
492 SSP1-1.9 significantly differ from those under the other two scenarios. However, all
493 scenarios are highly favorable for an increase in SSRD, suggesting that the potential
494 risk of high O_3 concentrations may be increasing in the forthcoming decades, even
495 taking the Green Road. Fortunately, based on recent emission inventories, pollutants in
496 China have shown a decreasing trend. In our study, by comparison with emissions in
497 the summer of 2021, VOCs and NO_x emissions in the summer of 2022 decreased by 4%
498 and 5%, respectively (Jiang et al., 2022; Li et al., 2024), leading to a reduction in O_3
499 concentration by 1.5 µg m^{-3}. According to these emission reduction rates, we use a
500 simple linear extrapolation method, also in conjunction with China's carbon neutrality
501 goal, to estimate VOCs and NO_x emissions in the future. By the 2030 carbon peak,
502 VOCs and NO_x emissions will have been reduced by approximately 31% and 37%,
503 respectively, relative to 2021 levels. By the 2060 carbon neutrality goal, the reductions
504 are projected to reach 80% and 87%, respectively. Actually, emission reductions may
505 face challenges and unlikely to follow such a perfect pathway, and the response of O_3
506 concentration to precursor reductions is also nonlinear. We thus assume that, if such an
507 idealized scenario is followed, O_3 concentrations by 2030 and 2060 are estimated to be
508 reduced by 13.5 µg m^{-3} and 58.5 µg m^{-3}, respectively, relative to the levels in 2021.
509 Therefore, in the long term, on the decadal scale, the continued emission reductions are
510 expected to significantly reduce O_3 concentration.
511
512 However, on an interannual scale, the projected SSRD variability can reach several tens
513 of W m^{-2}, which is consistent with this study. Our study shows that interannual
514 difference in SSRD between the summers of 2022 and 2021 is more than 80 W m^{-2}.
515 Based on the linear relationship between O_3 and SSRD shown in Figure 2, such
516 differences in SSRD corresponds to a change of 28 µg m^{-3} in daytime O_3 concentration.
517 According to the spatial distribution in Figure 4, the regional mean daytime O_3 change
518 due to meteorological changes (including clouds and SSRD) is 9.2 µg m^{-3}. Thus, a
519 sudden increase in SSRD may partially offset the benefits of emission reductions. Given
520 that coordinated VOCs and NO_x emission reductions are in the early stage, the

521 increasing possibility of highly favorable photochemical conditions under climate
522 change could not only counteract the effects of emission reductions, but may even lead
523 to a rebound in O_3 concentrations in the short term. Moreover, less clouds and more
524 SSRD under SSPs will also weaken CRI and consequently aggravate O_3 pollution in
525 the future. Based on SSPs projections, the interannual differences of summer SSRD
526 and cloud cover could reach or even exceed those observed between the summers of
527 2022 and 2021 (the interannual differences in SSRD and LCC is 82.7 W m^{-2} and 0.09,
528 respectively). It is reasonable to expect that the CRI interannual variability will likely
529 exert an influence on O_3 changes that is no less significant than the calculation
530 presented in this study. Though these factors related to climate change are highly
531 variable, based upon the past and present impacts on O_3 concentration, their impacts on
532 future O_3 pollution control are widely believable. Thus, we suggest that, if
533 anthropogenic emission reductions are insufficient, these changes in clouds and SSRD
534 linked to climate change will increase O_3 concentration during the warm season.
535
536 **4 Conclusions and implications**
537 High O_3 concentration during the warm season have been increasingly becoming a
538 major air pollution issue in China, however, whether it is closely connected to climate
539 change has not yet received sufficient attention. Our findings indicate that the sudden
540 increase in O_3 concentration in the YRD during the warm season of 2022 is closely
541 linked to the weak CRI characterized by lower LCC and higher SSRD. Less LCC favors
542 more solar shortwave radiation reaching the surface, which significantly accelerates
543 photochemistry, thereby leading to a pronounced increase in O_3 concentration.
544
545 The notable increase in O_3 concentration caused by weakened CRI has significantly
546 exceeded the O_3 reduction caused by the interannual decrease in precursor emissions
547 during the warm season of 2022, attenuating the benefits of precursor emission
548 reductions. We emphasize that the focus on LCC and SSRD is with significant
549 implications for operational forecast on O_3 pollution, i.e., more stringent measures on
550 precursor emission reductions are imperative under weaker CRI.
551
552 Our results suggest that climate warming will make O_3 pollution control more
553 challengeable via altering clouds and SSRD and weakening the CRI. The high-level O_3
554 is not only influenced by changes in clouds and solar radiation related with short-term
555 synoptic-scale circulation adjustments, but also modulated by long-term climate change.
556 The occurrence likelihood of heatwaves (Chen et al., 2019; Ma et al., 2023),
557 accompanied by fewer clouds and more SSRD, will increase under climate warming.
558 Furthermore, if anthropogenic emissions are not greatly reduced, human-induced
559 forcing will further amplify the probability (Faranda et al., 2023; King et al., 2016;
560 Lopez et al., 2018; Sun et al., 2017; Zhang et al., 2024). Inevitably, the co-occurrence

561   of extreme high temperature and O_3 concentration is likely to occur frequently, posing

562   a greater threat to human health, crops, and vegetation. Therefore, we would like to

563   propose that more proactive human actions are vital to offset the penalty of climate

564   change to these issues.

565

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853

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856   and suggestions on the structure and highlights of the paper. T.F. and M.D. revised the

857   manuscript. B.T., X.H., B.H., D.Y., and S.G. participated in data analysis and

858   discussions on the results.

859

860   **Competing interests:** The authors declare that they have no competing interests.

861

862   **Data availability:**

863   Hourly observation data on mass concentrations of ambient air pollutants at surface

864   released by the Ministry of Ecology and Environment, China are available on the

865   website of *https://www.aqistudy.cn*.

866   Monthly mean low cloud cover and downward solar radiation from European Center

867   for Medium-Range Weather Forecasts ERA5 reanalysis data are obtained by a

868   registration on the website of *https://cds.climate.copernicus.eu/datasets/reanalysis-*

869   *era5-single-levels-monthly-means?tab=download*.

870   Hourly observation data on meteorological parameters at weather stations are from

871   National Oceanic and Atmospheric Administration, available on the website of

872   *https://www.ncei.noaa.gov/maps/hourly*.

873   National Centers for Environmental Prediction Final Analyses data are from

874   *http://rda.ucar.edu/datasets/ds083.2*.

875   Initial and boundary chemical fields are available on the website of

876   *https://www.acom.ucar.edu/cam-chem/cam-chem.shtml*.

877   The MEIC emission inventory developed by Tsinghua University is from the website

878   of *http://meicmodel.org.cn*.

879   The projected total cloud cover percentage and surface downwelling shortwave

880   radiation are from *https://cds.climate.copernicus.eu/datasets/projections-*

881   *cmip6?tab=downloa*d.

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883 **Figures and Tables**

884 **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

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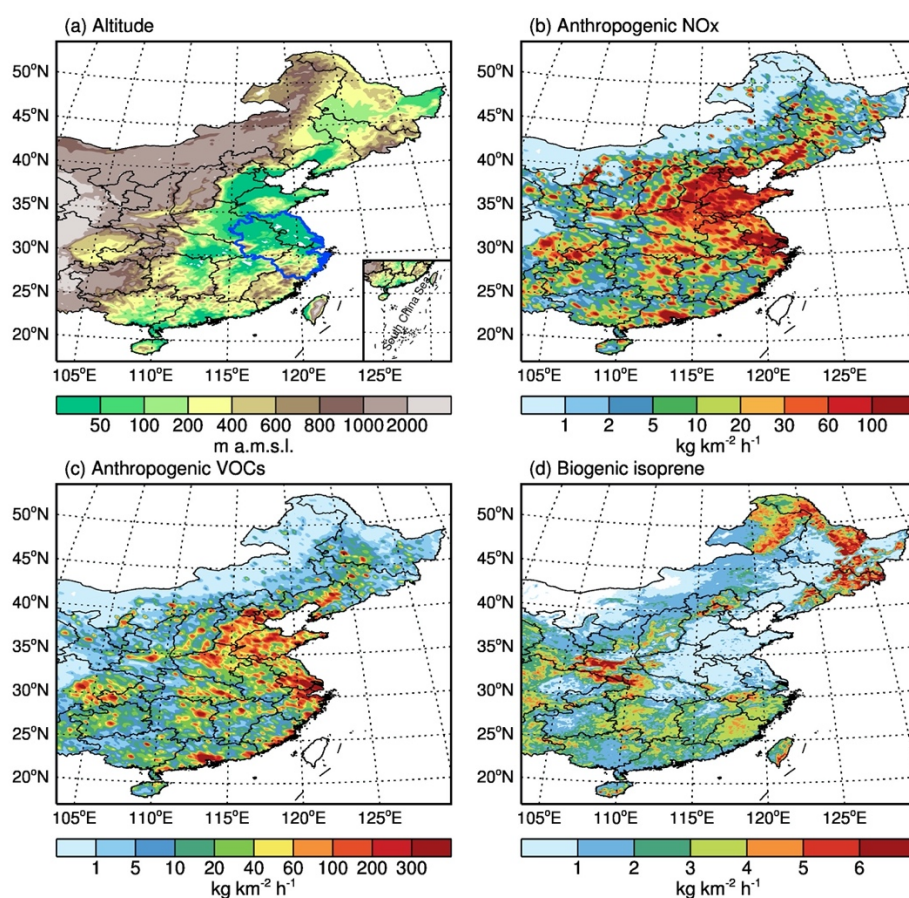
888   **Figure 1** Location of the YRD, China and spatial distributions of emissions. (a) The

889   flat YRD is located in the eastern China, marked by the blue line. (b) Anthropogenic

890   NO_x emission rate in July 2022 based on the MEIC emission inventory. (c) Same as (b),

891   but for anthropogenic non-methane VOCs. (b) and (c) represent human-induced

892   emissions of precursors for O_3. (d) Biogenic isoprene emission rate is calculated by

893   MAGAN, representing biogenic VOCs emissions.

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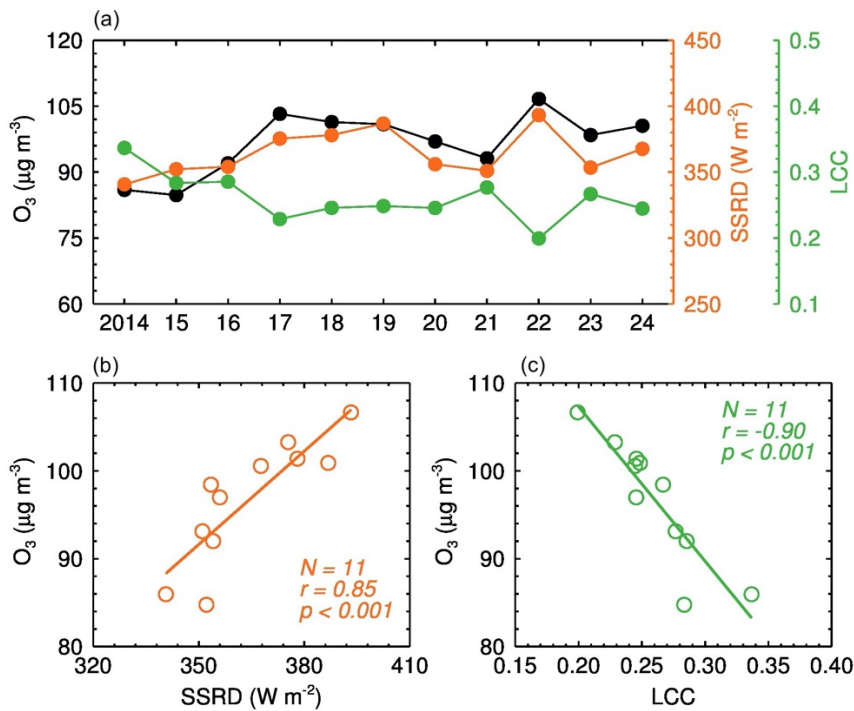
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897 **Figure 2** Observed relationships between O_3, SSRD, and LCC. (a) Annual variation in
898 mean daytime O_3 concentration (black), SSRD (orange), and LCC (green) during the
899 warm season of the past decade (2014-2024) in the YRD, China. (b) Correlation
900 between O_3 concentration and SSRD. (c) Correlation between O_3 concentration and
901 LCC. The colored lines in (b) and (c) represent the linear fits through the data in (a),
902 i.e., $[O_3] = -32.20 + 0.35 \times [SSRD]$ with $r = 0.85$ and $[O_3] = 142.49 -173.87 \times [LCC]$
903 with $r =- 0.90$. O_3 concentration is significantly positively (negatively) correlated with
904 SSRD (LCC), with confidence levels exceeding 99.9%. Sample sizes N, correlation
905 coefficients r, and confidence levels p by the Student's t-test are shown in (b) and (c).
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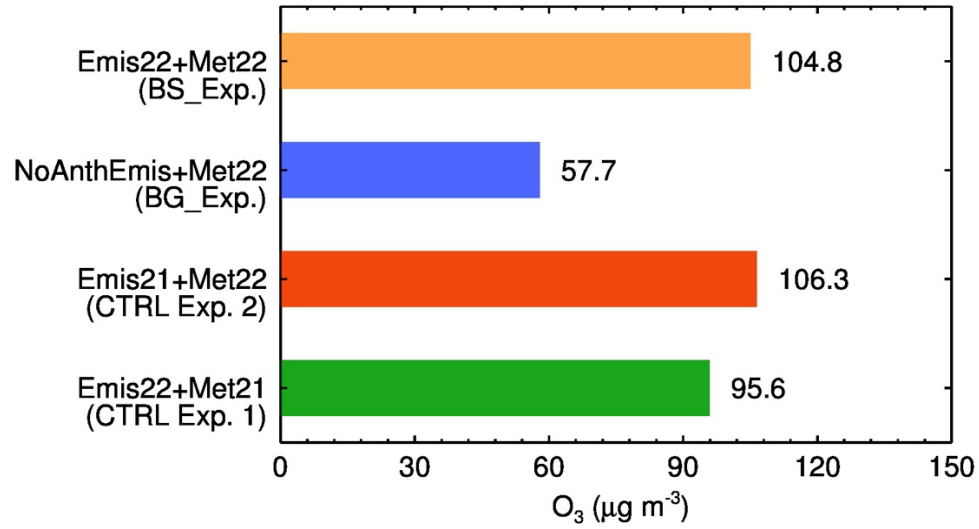
908 **Figure 3** Simulated O_3 concentrations under different model experiments.
909 Contributions of background input (blue) and anthropogenic emissions (orange *minus*
910 blue) to summer O_3 concentrations in the YRD. Contributions of emission change
911 (orange *minus* red) and meteorology change (orange *minus* green) to O_3 change.
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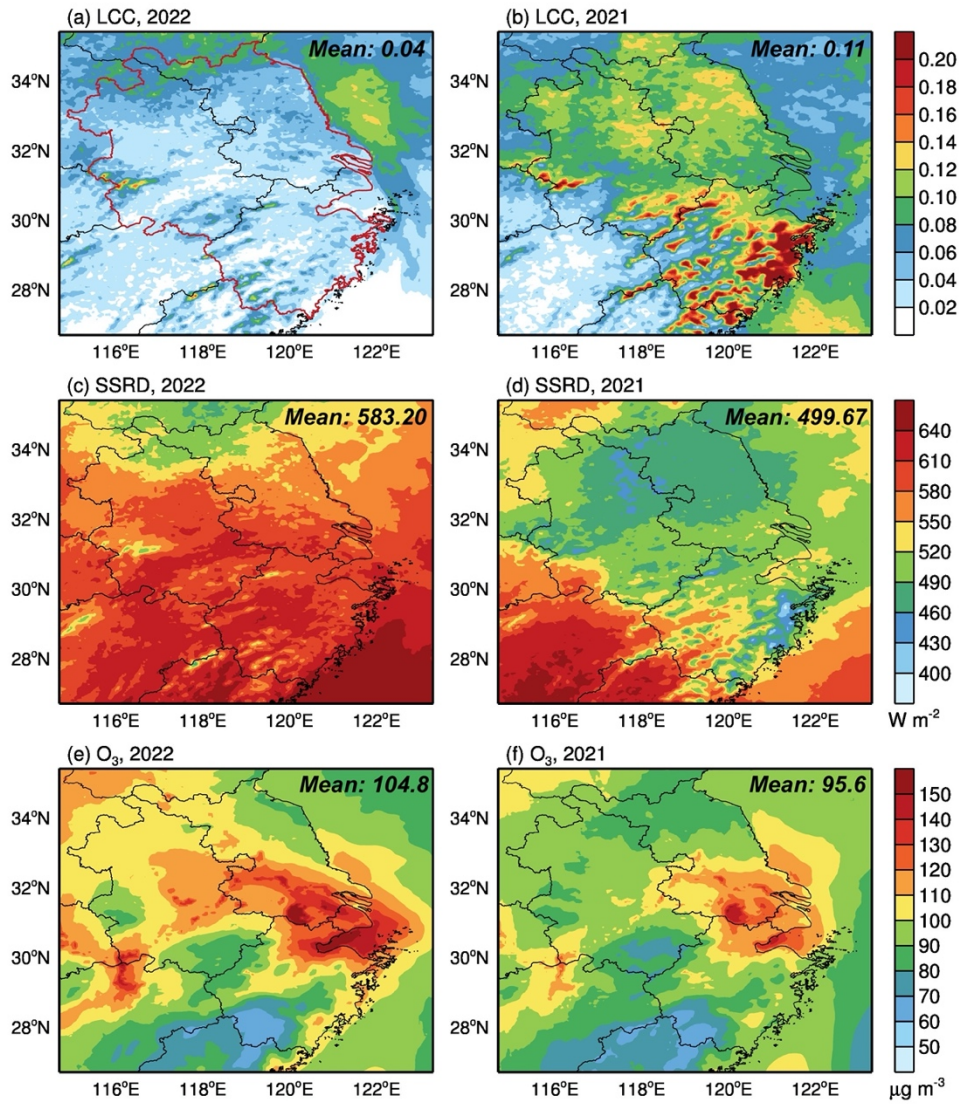
915 **Figure 4** Comparison of spatial distribution of monthly mean daytime O_3
916 concentrations under different CRI intensities. (a-b) LCC in July 2022 and July 2021,
917 respectively. (c-d) SSRD. (e-f) Daytime O_3 concentrations. (a) and (c) represent a weak
918 CRI mechanism due to less LCC and more SSRD, corresponding to higher O_3
919 concentrations, with a larger spatial coverage. (b) and (d) represent a strong CRI
920 mechanism due to more LCC and less SSRD, corresponding to lower O_3 concentrations,
921 with a smaller spatial coverage. The YRD is enclosed by the red line in (a). The regional
922 average of each variable is shown at the top-right corner of each panel.

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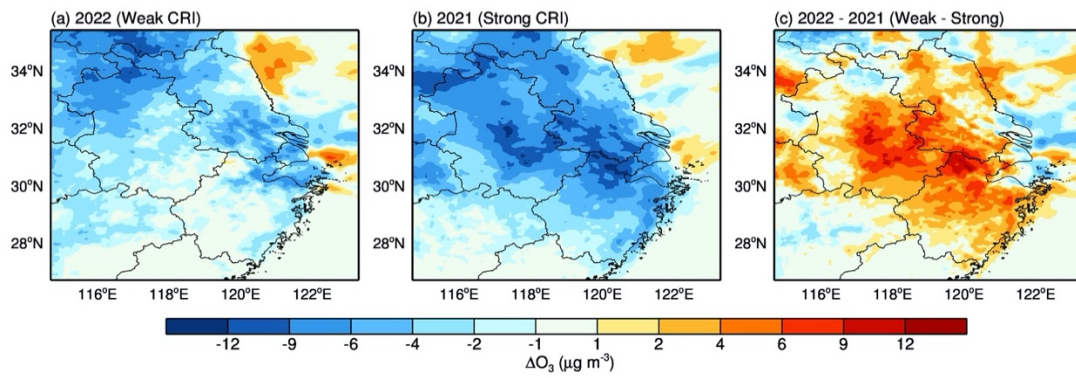
926 **Figure 5** Influence of CRI on O_3 change. (a) Difference in O_3 concentrations including
927 and excluding CRI in July 2022, indicating O_3 change caused by a weak CRI. (b) Same
928 as (a) but in July 2021, indicating O_3 change caused by a strong CRI. (c) A result of (a)
929 *minus* (b), representing ΔO_3 change caused by the CRI intensity change.
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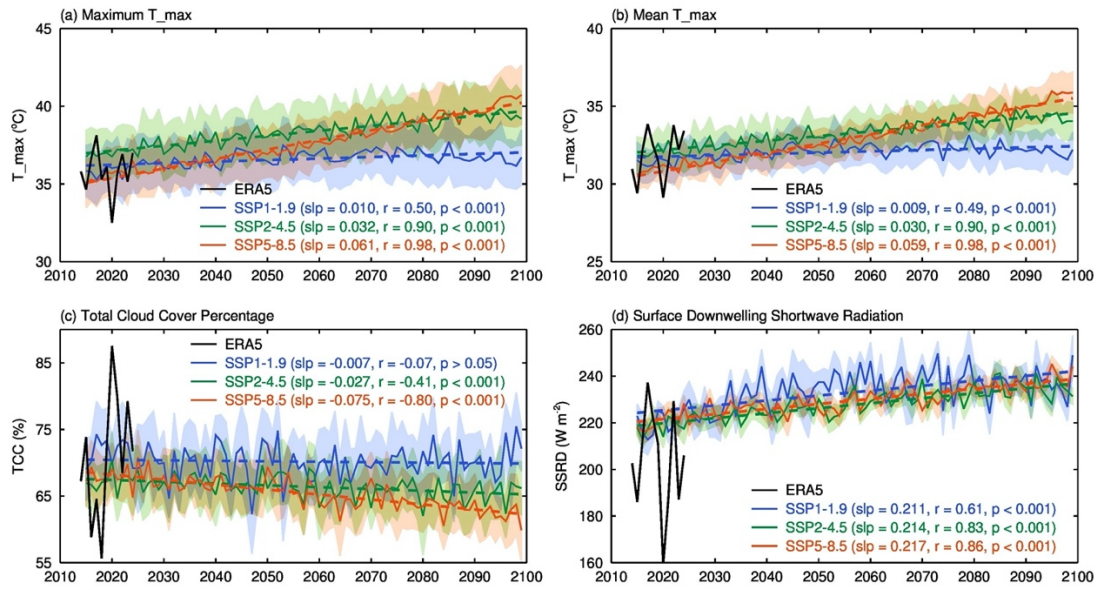
Figure 6 Trends of multi-model ensemble mean radiation conditions projected by the CMIP6 under three SSPs during 2015-2099. (a) The maximum daily T_max, and (b) The mean daily T_max in July, represent the extreme and mean status of high temperature, respectively. (c) TCC and (d) SSRD together reflect the solar radiation conditions for O_3 formation. The shading shows ±1.0 standard error, and the dash lines represent the linear trends of each variable under different SSPs. The black curve in each panel shows the real variation of each variable from the ERA5 reanalysis data during 2014-2024.