

1 **Investigating the response of China's surface ozone**  
2 **concentration to the future changes of multiple factors**

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14

## 15 **Abstract**

16 Climate change and associated human response are supposed to greatly alter  
17 surface ozone (O<sub>3</sub>), an air pollutant generated through photochemical reactions  
18 involving both anthropogenic and biogenic precursors. However, a comprehensive  
19 evaluation of China's O<sub>3</sub> response to these multiple changes has been lacking. We  
20 present a modelling framework under Shared Socioeconomic Pathways (SSP2-45),  
21 incorporating future changes in local and foreign anthropogenic emissions,  
22 meteorological conditions, and BVOCs emissions. From the 2020s to 2060s, daily  
23 maximum 8-hour average (MDA8) O<sub>3</sub> concentration is simulated to decline by 7.7 ppb  
24 in the warm season (April-September) and 1.1 ppb in non-warm season (October-  
25 March) over the country, with a substantial reduction in exceedances of national O<sub>3</sub>  
26 standards. Notably, O<sub>3</sub> decreases are more pronounced in developed regions such as  
27 BTH, YRD, and PRD during warm season, with reductions of 9.7, 14.8, and 12.5 ppb,  
28 respectively. Conversely, in non-warm season, the MDA8 O<sub>3</sub> in BTH and YRD will  
29 increase by 5.5 and 3.3 ppb, partly attributed to reduced NO<sub>x</sub> emissions and thereby  
30 weakened titration effect. O<sub>3</sub> pollution will thus expand into the non-warm season in  
31 the future. Sensitivity analyses reveal that local emission change will predominantly  
32 influence future O<sub>3</sub> distribution and magnitude, with contributions from other factors  
33 within ±25 %. Furthermore, the joint impact of multiple factors on O<sub>3</sub> reduction will be  
34 larger than the sum of individual factors, due to changes in the O<sub>3</sub> formation regime.  
35 This study highlights the necessity of region-specific emission control strategies to  
36 mitigate potential O<sub>3</sub> increases during non-warm season and under climate penalty.

## 37 **1 Introduction**

38 Surface ozone (O<sub>3</sub>) is a secondary air pollutant generated by photochemical  
39 reactions in the presence of two main kinds of precursors NO<sub>x</sub> (NO<sub>x</sub>=NO+NO<sub>2</sub>) and  
40 volatile organic compounds (VOCs). It has been reported to be a non-negligible threat  
41 to both human health and crop yield, and also a short-lived climate forcer with

42 warming effect (Finlaysonpitts and Pitts, 1997; Jerrett et al., 2009; Avnery et al., 2011;  
43 Lelieveld et al., 2015; von Schneidemesser et al., 2015; Tai and Val Martin, 2017; Yin  
44 et al., 2020; Feng et al., 2022; Niu et al., 2022). Given the abundant emissions of  
45 anthropogenic NO<sub>x</sub> and VOCs, China has suffered from extremely high and  
46 continuously increasing O<sub>3</sub> pollution from 2013 to 2019 with the peak season daily  
47 maximum 8-hour average (MDA8) O<sub>3</sub> concentration over 95 μg m<sup>-3</sup>. The rising trend  
48 has been reversed since 2020, along with the national annual NO<sub>x</sub> and non-methane  
49 VOCs (NMVOCs) emissions reduced by 28.3 % and 3.8 %, respectively during 2013–  
50 2020 (Zheng et al., 2018; Xiao et al., 2022; Liu et al., 2023a; Wang et al., 2023).  
51 However, current O<sub>3</sub> concentration over China is still much higher than the global air  
52 quality guidelines (60 μg m<sup>-3</sup> for the averaged peak season MDA8 O<sub>3</sub>, WHO, 2021).  
53 This presents a great challenge for the country to meet the criteria for public health  
54 welfare in the future (Feng et al., 2023; Jiang et al., 2023).

55 In addition to the anthropogenic driver, studies also addressed the roles of  
56 meteorological factors, biogenic VOCs (BVOCs) emissions and transboundary  
57 transport of pollutants on O<sub>3</sub> enhancement in China (Monks et al., 2015; Lu et al., 2019;  
58 Cao et al., 2022; Wang et al., 2022; Weng et al., 2022; Jiang et al., 2023).  
59 Meteorological factors, including temperature, humidity, wind, etc., influence the  
60 chemical reactions associated with O<sub>3</sub> production and elimination, and the  
61 transportation of O<sub>3</sub> precursors (Gong and Liao, 2019). The changed meteorology was  
62 estimated to enhance the summer MDA8 O<sub>3</sub> concentration by 1.4 ppb yr<sup>-1</sup> during 2013–  
63 2019 in the North China Plain (NCP), nearly half of the overall O<sub>3</sub> growth of 3.3 ppb  
64 yr<sup>-1</sup> (Li et al., 2020a). BVOCs refer to VOCs emitted from terrestrial ecosystems and  
65 possess high reactivity in atmospheric chemical processes, mainly including isoprene,  
66 monoterpenes and sesquiterpene (Wu et al., 2020a). Cao et al. (2022) reported that  
67 BVOCs emissions in summer 2018 enhanced 8.6 ppb MDA8 O<sub>3</sub> averaged over China  
68 with the highest contribution over 30 ppb in southern China. Moreover, O<sub>3</sub> and its  
69 precursors could be transported over long distance, and transboundary foreign

70 anthropogenic emissions were estimated to contribute 2–11 ppb to near surface O<sub>3</sub> in  
71 China (Ni et al., 2018; Han et al., 2019).

72 In the context of future global change, substantial but uncertain changes will occur  
73 in economy, climate and land cover. According to the Sixth Assessment Report of  
74 Intergovernmental Panel on Climate Change (IPCC AR6 report), the global surface  
75 temperature will increase 0.2–3.7 °C till 2100 under different scenarios compared to  
76 2015 (IPCC, 2021, 2022). As a result, unfavorable meteorological extremes, such as  
77 high temperature extremes and ecological drought events, will be more frequent and  
78 intense (Hong et al., 2019; Porter and Heald, 2019; IPCC, 2021), leading to the  
79 deterioration of air quality named as climate penalty. To conquer the climate change  
80 and resulting air quality deterioration, a series of measures will be implemented, such  
81 as accelerating the transition to clean energy, upgrading industrial production  
82 technologies and strengthening pollution control measures. Attributable to these  
83 changes, annual mean surface O<sub>3</sub> in East Asia was projected to change by –13.9–6.1  
84 ppb till 2100 compared to 2015 (IPCC, 2021). However, limited by the coarse  
85 resolution of earth system model and the lack of consideration of the regional measures  
86 for reducing air pollution and carbon emissions, global estimation is insufficient for  
87 understanding how O<sub>3</sub> pollution in China will respond to the complex future change.

88 There have been some studies on how the above-mentioned changes will affect  
89 future O<sub>3</sub> level in China. Hong et al. (2019) reported that the 1-hour maximum O<sub>3</sub> in  
90 April to September will be enhanced by 2–8 ppb within large areas of China under  
91 RCP4.5 (representative concentration pathways 4.5, van Vuuren et al., 2011) scenario  
92 from the 2010s to 2050s. Under high-forcing scenarios, Li et al. (2023) projected the  
93 climate-driven O<sub>3</sub> concentration in the 2100s and found that O<sub>3</sub> concentration in  
94 southeast China would increase 5–20 % compared to the 2020s by a machine learning  
95 method. A warming climate should enhance the O<sub>3</sub> level, given the increasing  
96 frequency of atmospheric stagnation and heat waves (Hong et al., 2019; Wang et al.,  
97 2022; Gao et al., 2023; Li et al., 2023). The effect of anthropogenic emission change

98 on China's O<sub>3</sub> level has been estimated by studies under different scenarios. Zhu and  
99 Liao (2016) applied global emission estimates under RCPs, and found that the  
100 maximum growth of annual mean O<sub>3</sub> would be 6–12 ppb during 2000–2050 under  
101 different scenarios. Using the Dynamic Projection model for Emissions in China  
102 (DPEC) that better includes local information of energy transition and emission  
103 controls (Cheng et al., 2021b), Xu et al. (2022) reported that the joint impact of climate  
104 change and emission reduction would reduce the annual MDA8 O<sub>3</sub> concentration to  
105 63.0 µg m<sup>-3</sup> under ambitious scenario of carbon neutrality. Biogenic emission change  
106 is another factor influencing future O<sub>3</sub> (Chen et al., 2009; Andersson and Engardt, 2010;  
107 Harper and Unger, 2018; Wang et al., 2020). Liu et al. (2019) predicted a 24 % growth  
108 of BVOCs emissions driven by climate change under RCP8.5 from 2015 to the 2050s,  
109 resulting in a variation of daily 1-h maximum O<sub>3</sub> concentration ranging from –10.0 to  
110 19.7 ppb across different regions in China.

111 Limitations exist in current studies, which prevent comprehensive assessment and  
112 understanding of the joint impacts of future changes of multiple factors on China's O<sub>3</sub>  
113 pollution. Firstly, the above estimations mainly focused on the influence of future  
114 changes on summertime or annual average O<sub>3</sub> concentration. As China's O<sub>3</sub> pollution  
115 has been reported to spread into spring and fall, it is of great importance to separate the  
116 impacts on warm (April to September, the six months with heaviest O<sub>3</sub> pollution for  
117 most part of China, Liu et al., 2023a) and non-warm season O<sub>3</sub> (October to March),  
118 considering the diverse air pollution sources and O<sub>3</sub> formation sensitivity to precursors  
119 for different seasons (Li et al., 2021; Wang et al., 2023). Recent studies have suggested  
120 diverse effects of future emission change on O<sub>3</sub> evolution for difference seasons in  
121 China (Hou et al. 2023; Liu et al. 2023b). In addition, the rising frequency of extreme  
122 weathers and declining anthropogenic emissions will further influence the possibility  
123 of extreme O<sub>3</sub> events, which has been scarcely discussed. Secondly, to restrain global  
124 warming, China has made a national commitment to achieving “carbon neutrality” by  
125 2060 (Shi et al., 2021), and accordingly launched a series of energy and climate action

126 plans to reduce greenhouse gas emissions. These actions will also cause substantial  
127 reductions in air pollutant emissions, but have not been fully included in existing  
128 predictions of global emissions (Tong et al., 2020; Cheng et al., 2021b). Large bias will  
129 then be caused in the simulation of anthropogenic-induced future changes of air quality,  
130 with a less realistic estimate of local emission path (Cheng et al., 2021a). Due to  
131 probably faster decline of emissions in China but slower in surrounding countries in the  
132 future, the contributions of transboundary emissions on China's O<sub>3</sub> can be greatly  
133 changed and has not yet been fully considered (Hou et al., 2023). Thirdly, BVOCs  
134 emissions will not only be affected by meteorological factors but also by land use and  
135 land cover change (Penuelas and Staudt, 2010; Szogs et al., 2017; Wang et al., 2021a).  
136 Future land management will change due to socio-economic development and  
137 necessary actions as climate change response, and the changed shares of forest,  
138 cropland and grassland will alter the magnitude and distribution of BVOCs emissions  
139 and thereby affect O<sub>3</sub> concentration (Hurt et al., 2020; Liao et al., 2020; Liu et al.,  
140 2022). Finally, the existing evaluations were conducted separately for individual  
141 influencing factors, with diverse methods and data. The interactions between different  
142 factors were seldom included in existing analyses, and the relative contributions of  
143 multiple factors were difficult to be evaluated or compared. Relevant studies have been  
144 conducted in developed countries (Gonzalez-Abraham et al., 2015), and are still lack in  
145 China.

146 In this study, we evaluate the complex influence of future changes of multiple  
147 factors on surface O<sub>3</sub> concentration in China within a uniform framework. The  
148 evaluation is conducted from the perspectives of seasonal, regional and extreme events  
149 of O<sub>3</sub> pollution. Four factors are included in the analyses, i.e., meteorological conditions,  
150 local anthropogenic emissions, BVOCs emissions, and anthropogenic emissions from  
151 surrounding foreign countries. The analyses are conducted based on a series of  
152 sensitivity experiments in numerical modelling of future air quality, and up-to-date  
153 input data from multiple sources are utilized in the model (see details in next section).

154 We provide a comprehensive perspective on the spatiotemporal change of China's O<sub>3</sub>  
155 pollution till the 2060s, under a moderate way SSP2 of Shared Socioeconomic  
156 Pathways (SSPs, Riahi et al., 2017) and a midrange mitigation scenario RCP4.5, a  
157 scenario at the middle of the socio-economic developing way with radiative forcing at  
158 4.5 W m<sup>-2</sup> nominally by 2100 (Meinshausen et al., 2020). The outcomes highlight the  
159 regional and seasonal heterogeneity of O<sub>3</sub> pollution risks driven by complex future  
160 change of multiple factors, and support strategy design of O<sub>3</sub> pollution alleviation with  
161 specific principles, targets and action pathways.

## 162 **2 Data and Methods**

### 163 **2.1 Main framework and research domain**

164 The simulation framework incorporates the Weather Research and Forecasting  
165 model (WRF, version 3.7.1) to generate hourly meteorological fields, the Model of  
166 Emissions of Gases and Aerosols from Nature (MEGAN, version 2.1) to calculate  
167 gridded BVOCs emissions, and the Community Multiscale Air Quality model (CMAQ,  
168 version 5.2) to simulate O<sub>3</sub> concentration. BVOCs emission calculations and air quality  
169 simulations are driven by meteorological fields of 2018–2022 (the 2020s, representing  
170 the current situation) and 2058–2062 (the 2060s, representing the future situation). All  
171 simulation results are averaged over a period of five years to mitigate the influence of  
172 interannual variability of meteorology. The modelling domain, same for WRF,  
173 MEGAN and CMAQ, covers East Asia, most areas of South Asia and Central Asia, and  
174 part of Southeast Asia and North Asia (Figure 1). It applies the Lambert Conformal  
175 Conic projection centered at (110° E, 34° N), and the horizontal resolution is 27 km×27  
176 km, with 303×203 grids. The target area, Chinese mainland, includes 31 provincial-  
177 level administrative regions (excluding Hong Kong, Macao and Taiwan). Eight  
178 geographical regions are defined, and locations of the three regions with dense  
179 population and relatively heavy air pollution are also shown in Figure 1, namely BTH  
180 (Beijing-Tianjin-Hebei), YRD (Yangtze River Delta) and PRD (Pearl River Delta).

## 181 2.2 Data sources and processing methods

182 We use the bias-corrected RCP4.5 output of the National Center for Atmospheric  
183 Research's Community Earth System Model (NCAR CESM) as initial and boundary  
184 conditions for WRF (Monaghan et al., 2014). A ten-year dynamic downscaling  
185 simulation for 2018–2022 and 2058–2062 is conducted. Note we do not utilize the real-  
186 time reanalysis data to drive the simulation of the 2020s, in order to minimize the  
187 systematic error between the simulation driven by real meteorological conditions (for  
188 current simulations) and climate projection (for future simulations),

189 The BVOCs emissions are basically determined by meteorology and vegetation.  
190 The meteorological conditions are supplied by WRF. The vegetation data, including  
191 leaf area index (LAI), plant functional types (PFTs) and emission factors (EFs) of each  
192 PFT, are determined for 2020 and 2060. Gridded LAI data for 2020 are obtained from  
193 Global Land Surface Satellite product (Liang et al., 2021), and those for 2060 under  
194 SSP2-45 scenario are downscaled from the daily CESM2 output of Coupled Model  
195 Intercomparison Project Phase 6 (CMIP6). PFTs data for 2020 are derived from  
196 MCD12C1 product of Moderate-Resolution Imaging Spectroradiometer (MODIS)  
197 dataset and mapped to the 16 types required for MEGAN following Liao et al. (2020).  
198 The PFTs data for 2060 in China are obtained from Liao et al. (2020) under SSP2-45  
199 scenario, with other regions maintaining those of 2020. EFs for each PFT are taken  
200 from Guenther et al. (2012).

201 Anthropogenic emissions for Chinese mainland are obtained from the Multi-  
202 resolution Emission Inventory for China (MEIC,  
203 [http://meicmodel.org.cn/?page\\_id=560](http://meicmodel.org.cn/?page_id=560)) for 2020, and DPEC version 1.1 under SSP2-  
204 45 incorporating the best available end-of-pipe pollution control technologies for 2060.  
205 The annual total emissions by country/region outside Chinese mainland are obtained  
206 from CMIP6 dataset under SSP2-45 scenario (O'Neill et al., 2016; Gidden et al., 2019),  
207 and these emissions are downscaled into gridded monthly data for CMAQ simulation,  
208 based on the spatial and temporal distributions of emissions in MIX Asian emission



209 inventory (Li et al., 2017). The speciation profiles of NMVOCs are taken from MIX as  
210 well. Supplementary Figure S1 shows the emissions of two main precursors of O<sub>3</sub> by  
211 year and region. The NO<sub>x</sub> and NMVOCs emissions for Chinese mainland were  
212 estimated to decline 58 % and 51 % from 2020 to 2060, respectively, much faster than  
213 those of surrounding areas within the modelling domain (8 % and 14 % respectively).  
214 In particular, the NO<sub>x</sub> emissions would decline 57–62 % for the three developed regions  
215 BTH, YRD and PRD, while the reductions of anthropogenic NMVOCs would vary a  
216 lot among regions (36 %, 49 % and 60 % for BTH, YRD and PRD, respectively).

217 Carbon Bond 2005 (CB05, Yarwood et al., 2005) is adopted as the gas-phase  
218 chemical mechanism and the sixth-generation CMAQ aerosol module AERO6 (Appel  
219 et al., 2013) as aerosol chemistry mechanism. The initial and boundary conditions are  
220 set by default clean air conditions in CMAQ, and the first 10 days for each year are  
221 determined as the spin-up period to minimize the effects of initial and boundary  
222 conditions.

### 223 **2.3 Simulation cases**

224 Six cases of CMAQ simulations are conducted to investigate the impacts of future  
225 change of the four factors on O<sub>3</sub> concentration in China (Table 1). Cases 1 and 2  
226 represent the current (2020s) and future (2060s) baseline, respectively, and the  
227 difference between them indicates the joint effect of the future changes of multiple  
228 factors. Each of Cases 3–6 applies the prediction for 2060s for one specific factor but  
229 keeps the remaining factors at current condition (2020s). Thus, the difference between  
230 each of those four cases and Case 1 indicates the impact of individual factor, including  
231 meteorological conditions (Case 3), domestic anthropogenic emissions (Case 4),  
232 BVOCs emissions (Case 5) and anthropogenic emissions of surrounding countries  
233 (countries other than Chinese mainland within the modeling domain, Case 6). Each case  
234 contains a five-year (2018–2022 or 2058–2062) WRF-MEGAN-CMAQ simulation  
235 driven by the varying meteorological conditions for individual years, and the five-year  
236 average of simulated O<sub>3</sub> concentrations is adopted for further analyses.

## 237 **2.4 Model performance**

238 To evaluate the model performance, we conduct a comparative analysis between  
239 simulations and observations for meteorological factors and O<sub>3</sub> concentrations, as well  
240 as an intercomparison for BVOCs estimates between different studies.

241 We first examine the capability of downscaled CESM climate projections in  
242 capturing the meteorological conditions of the 2020s. We applied the meteorological  
243 data from the National Climate Data Center (NCDC, archived at <https://quoteoft.net/air>)  
244 in 2020, and the statistical metrics are presented in Supplementary Table S1. The  
245 modeled temperature at 2 m (T<sub>2</sub>) is in good spatiotemporal agreement with the  
246 observations, with the correlation coefficient (R) of 0.96 and index of agreement (IOA)  
247 of 0.98. The relative humidity (RH) is also well predicted with R and IOA at 0.78 and  
248 0.88, respectively. The model shows an overestimation on the wind speed by 1.41 m  
249 s<sup>-1</sup>, which is also reported by Hu et al., (2022). The correlation coefficients of wind  
250 speed and direction are higher than 0.5. Overall, the modeled meteorological fields have  
251 basically captured the conditions in China and are appropriate for subsequent MEGAN  
252 and CMAQ simulations.

253 For BVOCs emissions, we compare our estimates for the 2020s with previous  
254 studies, as summarized in Supplementary Table S2. The total BVOCs, isoprene and  
255 terpenes emissions in this study are estimated at 33.55, 21.08 and 3.30 Tg yr<sup>-1</sup>,  
256 respectively, and are comparable to other studies. In particular, our estimate is larger  
257 than others except for Li et al. (2020b) for isoprene, while smaller than others except  
258 for Wu et al. (2020a) for terpenes. The differences between studies might result from  
259 the diverse strategies of mapping PFTs from the original satellite products and the  
260 difference between downscaled climate conditions and the real meteorological fields.

261 We apply the observed MDA8 O<sub>3</sub> concentration data from the national network of  
262 China Ministry of Ecology and Environment (archived at <https://quoteoft.net/air>) to  
263 evaluate CMAQ performance. As shown in Supplementary Figure S2, the simulation  
264 could capture the spatiotemporal distribution of surface MDA8 O<sub>3</sub> concentration for the

265 whole country and specific O<sub>3</sub> pollution hot spots, e.g., BTH and eastern Sichuan  
266 province with their surrounding areas. The statistical metrics of the comparisons  
267 between the simulated and observed monthly average MDA8 O<sub>3</sub> concentration of 2020s  
268 are summarize in Supplementary Table S1. The normalized mean biases (NMB) are  
269 calculated at 14.12 % and 10.90 % for warm and non-warm season, and R values at  
270 0.71 and 0.32, respectively. Even with a slight overestimation, the reliability of our  
271 simulation is comparable to most previous studies in China, with a better performance  
272 in the warm season (Hu et al., 2016; Lu et al., 2019; Gao et al., 2020; Yang and Zhao,  
273 2023).

274 We evaluate the interannual variability within each of the five-year simulations,  
275 based on the coefficient of variation (CV), the ratio of standard deviation to mean of  
276 the simulated O<sub>3</sub> concentration. As shown in Supplementary Table S3, the CVs are  
277 generally below 5 % in most cases, indicating relatively small interannual variability in  
278 O<sub>3</sub> concentration simulation. The results thus justify the representativeness of the five-  
279 year averages for present and future scenarios.

## 280 **3 Results and Discussions**

### 281 **3.1 Future change of meteorology and BVOCs emissions**

282 The downscaled changes in the meteorological factors from the 2020s to 2060s  
283 (SSP2-45 scenario) are shown in Figure 2, including temperature, RH and wind speed  
284 (WS). The changes are analyzed separately for April–September (warm season) and  
285 October–March (non-warm season). For the warm season, daily maximum temperature  
286 at 2 m (T-max) will increase across China with an average change of 1.0 °C, and the  
287 minimum and maximum changes are found in Tibetan Plateau at 0.1 °C and in  
288 Heilongjiang province at 2.1 °C, respectively. The RH will decrease slightly by -0.6 %  
289 for the whole country, with the changes for most areas within the range between -3 %  
290 and 0 % except for some areas of Northwestern China, Southwestern China, and  
291 Tibetan Plateau (see the region definitions in Figure 1). The growing T-max and

292 declining RH will enhance the photochemical production of O<sub>3</sub> and BVOCs emissions.  
293 For the non-warm season, the national average growth of T-max will be smaller at 0.2 °C  
294 and some areas in Northeastern, Northern and Eastern China will even experience a  
295 decline ranging from -1.8 to 0 °C. The RH will change diversely across the country,  
296 ranging from -6.0 to 6.3 %. Very limited change in WS will occur, ranging from -0.1  
297 to 0.2 m s<sup>-1</sup> in most areas of the country. Generally, the decreasing wind speed in future  
298 East Asia could be attributed to weakened atmospheric circulation (Coumou et al., 2018;  
299 Deng et al., 2021). The increasing wind speed in non-warm season might result from  
300 the temperature and pressure gradients between the land and adjacent oceans (Yao et  
301 al., 2019; Wu et al., 2020b). The spatial distribution of downscaled future  
302 meteorological field changes is generally in agreement with those predicted by Hong et  
303 al. (2019) and Hu et al. (2022). Some discrepancies in temperature and wind speed  
304 change of non-warm season between studies result from the different choices of base  
305 year and parameterization schemes of WRF.

306 Table 2 shows China's BVOCs emissions of the 2020s and 2060s (SSP2-45  
307 scenario) estimated with MEGAN, as well as the BVOCs emission intensity (emissions  
308 per unit area) for the three developed regions. The emissions will increase from 33.6  
309 Tg yr<sup>-1</sup> for the 2020s to 43.4 Tg yr<sup>-1</sup> for the 2060s. The growth rates in BTH, YRD and  
310 PRD are predicted to be 21.4 %, 23.9 % and 23.0 %, respectively, smaller than that for  
311 the whole country (29.2 %). The spatial distributions of BVOCs emissions for the 2020s  
312 and the changes from the 2020s to 2060s, are shown in Supplementary Figure S3. Areas  
313 all over China will experience the growth of BVOCs emissions, and it will be more  
314 prominent in areas with high vegetation coverage (e.g., Southern and Southwestern  
315 China) rather than urban areas. The growth of BVOCs emissions will enhance the  
316 contribution of natural sources to O<sub>3</sub> formation, especially along with declining  
317 anthropogenic emissions in the future (Penuelas and Llusia, 2003; Riahi et al., 2017;  
318 Gao et al., 2022).

### 319 **3.2 Response of surface O<sub>3</sub> concentration to combined future changes**

320 Figure 3 illustrates the spatial distributions of MDA8 O<sub>3</sub> concentrations for the  
321 warm and non-warm seasons of the 2020s and 2060s (SSP2-45 scenario), as well as the  
322 differences between the two periods. Briefly, future changes of the four factors under  
323 SSP2-45 are estimated to jointly reduce MDA8 O<sub>3</sub> by 7.7 and 1.1 ppb in the warm and  
324 non-warm season, respectively, while the O<sub>3</sub> responses to future changes will differ by  
325 region.

326 In the warm season of the 2020s (Figure 3a), the nationwide average MDA8 O<sub>3</sub>  
327 concentration is simulated at 57.3 ppb, and those of BTH, YRD and PRD are 73.7, 68.7  
328 and 52.3 ppb, respectively. Hot spots of O<sub>3</sub> pollution, with average MDA8 O<sub>3</sub> over 75  
329 ppb, are mainly located in Northern China and Sichuan province. The pattern is  
330 predicted to persist into the 2060s (Figure 3b), with a decline in both the severity and  
331 size of highly polluted regions. The nationwide MDA8 O<sub>3</sub> concentration will decline  
332 13.4 % to 49.6 ppb, and that in most areas of China will be within the range of 37.5–  
333 67.5 ppb. The highest concentration will be lower than 75 ppb for the two hotspots of  
334 Northern China and Sichuan. BTH will remain as the most O<sub>3</sub>-polluted area in warm  
335 season, with the O<sub>3</sub> concentration at 63.9 ppb (13.3 % smaller than the 2020s), while  
336 that of YRD and PRD will decrease to 53.9 (21.5 %) and 39.8 ppb (23.9 %),  
337 respectively. O<sub>3</sub> concentration in the developed regions will decline faster than or  
338 roughly the same as that for the whole country. The reductions in MDA8 O<sub>3</sub> from 2020s  
339 to 2060s will be 10–20 ppb for Northern, Eastern, Central and Southern China and 0–  
340 10 ppb for Northeastern and Northwestern China as well as the Tibetan Plateau (Figure  
341 3c). Notably, some areas in Sichuan are expected to experience a substantial decline of  
342 MDA8 O<sub>3</sub> over 20 ppb.

343 O<sub>3</sub> concentration of the non-warm season is simulated to be much lower than that  
344 of the warm season. The 2020s average MDA8 O<sub>3</sub> is 48.4 ppb, ranging from 30.0 to  
345 67.5 ppb in most areas of China (Figure 3d). Different from the warm season in which  
346 highest concentration is found for Northern China and Sichuan, the Southern and  
347 Southwestern parts of China suffer the highest O<sub>3</sub> level for the non-warm season. A

348 general west-to-east and south-to-north gradient is found for MDA8 O<sub>3</sub>, with the lowest  
349 concentration found in Northern and Northeastern China. The concentrations in BTH  
350 and YRD are simulated at 33.8 and 45.1 ppb, respectively, much lower than that of  
351 PRD (58.9 ppb). Relatively high temperature during even the non-warm season is  
352 expected to expand the O<sub>3</sub> pollution period in Southern China. Resulting from complex  
353 change of multiple factors, the national average MDA8 O<sub>3</sub> concentration in the non-  
354 warm season of 2060s will decrease slightly to 47.3 ppb under SSP2-45, and that in  
355 most regions will be within the range of 37.5–52.5 ppb except for some areas in  
356 Northeastern China and Tibetan Plateau (Figure 3e). The MDA8 O<sub>3</sub> concentrations of  
357 the three developed regions will become closer at 39.3, 48.4 and 51.6 ppb for BTH,  
358 YRD and PRD, respectively. As illustrated in Figure 3f, MDA8 O<sub>3</sub> is predicted to  
359 increase in BTH and YRD and the surrounding areas, with the growth mostly ranging  
360 0–15 ppb. In other areas (especially in Southern China), the concentration will decrease  
361 in the non-warm season by –15 to –5 ppb. As a result of the increased O<sub>3</sub> in the less  
362 polluted Eastern and Northern China and decreased O<sub>3</sub> in the more polluted  
363 Southwestern and Southern parts, the 2060s regional disparity in the non-warm season  
364 O<sub>3</sub> pollution will get smaller compared to the 2020s (Figure 3d and 3e).

365 To further explore the temporal pattern of O<sub>3</sub> level in the future, we compare the  
366 monthly average MDA8 O<sub>3</sub> in the 2020s and 2060s under SSP2-45 for the whole  
367 country and three developed regions (Figure 4 and Supplementary Figure S4). For the  
368 whole country (Figure 4a), the changes of monthly average MDA8 O<sub>3</sub> from 2020s to  
369 2060s are estimated to range from –3.2 to –10.7 ppb in the warm season but less  
370 prominent in the non-warm season (from –2.7 to 0.9 ppb). Along with the more  
371 reduction in summertime (June, July and August), in particular, the periods with the  
372 highest O<sub>3</sub> concentration will expand into spring (March) and fall (October), as  
373 presented in Supplementary Figure S4. For the three regions, a greater decline in O<sub>3</sub>  
374 concentration is found in the warm season while a smaller or even a growth is found in  
375 the non-warm season. For BTH (Figure 4b), the monthly MDA8 O<sub>3</sub> concentrations

376 range between 24.7 and 88.4 ppb in the 2020s with a clear difference between the warm  
377 and non-warm season. This pattern will remain in the 2060s with smaller difference  
378 between months (30.6–70.2 ppb). The temporal change pattern of YRD is similar to  
379 that in BTH, with decline in the warm season and growth in the non-warm season  
380 (Figure 4c). The shift of O<sub>3</sub> pollution from the warm towards the non-warm season is  
381 more prominent in the PRD, the only region where O<sub>3</sub> concentration of all the months  
382 in 2060s is predicted to decline (Figure 4d). Different from BTH and YRD, as  
383 mentioned above, higher O<sub>3</sub> concentrations during spring and autumn and lower in  
384 summer (due to the abundant summertime precipitation and high humidity) are found  
385 for PRD in the 2020s (Gao et al., 2020; Han et al., 2020). With great O<sub>3</sub> decline in the  
386 warm season, the periods experiencing peak O<sub>3</sub> pollution are predicted in the non-warm  
387 season of the 2060s, predominantly between October and March (Supplementary  
388 Figure S4).

### 389 **3.3 Identifying surface O<sub>3</sub> response to individual factors**

#### 390 **3.3.1 Local anthropogenic emission change**

391 Figure 5 shows the influences of changes of each individual factors (local  
392 anthropogenic emissions, meteorological conditions, BVOCs emissions, and  
393 anthropogenic emissions from surrounding countries) on the warm and non-warm  
394 season O<sub>3</sub> concentrations. Out of the four, the change of local anthropogenic emissions  
395 is predicted to be the most influential factor, resulting in a national average decline of  
396 7.2 and 0.8 ppb for the warm and non-warm season, respectively (Figure 5a and 5e). In  
397 the warm season, the emission reduction will play a positive role in reducing O<sub>3</sub>  
398 pollution in most areas of China, and the decrease will exceed 10 ppb across Northern,  
399 Eastern, Central, Southern and part of Southwestern China. In the non-warm season,  
400 emission reduction will have contrasting effects on MDA8 O<sub>3</sub> levels in the north and  
401 south part of China, enhancing MDA8 O<sub>3</sub> by 0–15 ppb for the former while restraining  
402 it by 0–10 ppb for the latter. Especially, the emission reduction is predicted to elevate

403 the O<sub>3</sub> concentration by 5.9 and 4.0 ppb for BTH and YRD respectively.

404 Supplementary Figure S5 shows the relative emission reductions from 2020s to  
405 2060s by region. Under SSP2-45 scenario, the reductions of NO<sub>x</sub> and VOCs emissions  
406 will range from 35.6 % to 63.6 % for different regions, and VOCs emission reduction  
407 will be less than that of NO<sub>x</sub> except for PRD. As the NO<sub>x</sub>-limited regime for O<sub>3</sub>  
408 formation (i.e., O<sub>3</sub> is more sensitive to NO<sub>x</sub> emission change) occurs more frequently  
409 in the warm season while the VOC-limited regime more in the non-warm season, the  
410 larger decline of NO<sub>x</sub> emissions than VOCs should be more effective in restraining the  
411 warm season O<sub>3</sub> pollution but has less benefit or even negative effect in the non-warm  
412 season (Sillman and He, 2002). Wintertime of NCP and YRD have been reported under  
413 the VOC-limited regime and the excessive NO<sub>x</sub> emissions play an important role in  
414 removing O<sub>3</sub> by titration effect (Jin and Holloway, 2015; Li et al., 2021; Wang et al.,  
415 2021b). This may explain the MDA8 O<sub>3</sub> increase during the non-warm season with  
416 insufficient reduction of VOCs (35.6 % and 49.5 %) but sharp reduction of NO<sub>x</sub> of 53.4 %  
417 and 60.3 % for NCP and YRD, respectively. Similarly, Hou et al. (2023) and Liu et al.  
418 (2023b) also predicted a growth of O<sub>3</sub> concentration in non-warm season over BTH and  
419 YRD under a net-zero carbon emission scenario, resulting from a weakened titration  
420 effect. Supplementary Figure S6 shows the monthly variation of O<sub>3</sub> and odd oxygen  
421 (O<sub>x</sub>, O<sub>x</sub>=O<sub>3</sub>+NO<sub>2</sub>, representing the real photochemical production potential of O<sub>3</sub>  
422 considering the titration effect) in the 2020s and 2060s. It should be noted that the  
423 growth of O<sub>3</sub> in the non-warm season in 2060s for BTH and YRD will be accompanied  
424 by minimal change of O<sub>x</sub>, while the declines of O<sub>3</sub> and O<sub>x</sub> will appear simultaneously  
425 in the warm season for the three regions and in non-warm season for PRD. This  
426 indicates that the growth of non-warm season O<sub>3</sub> in BTH and YRD should result partly  
427 from NO<sub>x</sub> reduction and thereby weakened NO titration, as titration is a key pathway  
428 of O<sub>3</sub> loss when the chemical reactivity is relatively low in winter (Gao et al., 2013;  
429 Akimoto and Tanimoto, 2022). The differentiated O<sub>3</sub> responses to precursor reduction  
430 between YRD and PRD have also been detected during the COVID-19 breakout period.



431 With the O<sub>3</sub> isopleth plots, Wang et al. (2021b) illustrated that 40–60 % reduction of  
432 NO<sub>x</sub> and VOCs enhanced the O<sub>3</sub> formation in YRD under the VOC-limited regime but  
433 suppressed O<sub>3</sub> in PRD under the transitional regime (a regime between NO<sub>x</sub>- and VOC-  
434 limited). Therefore, VOCs emission controls should be better addressed for O<sub>3</sub>  
435 pollution alleviation when it expands to non-warm season in the future.

### 436 **3.3.2 Meteorological condition change**

437 As shown in Figure 5b and 5f, the influence of meteorological change exhibits  
438 different patterns for the warm and non-warm season. In the warm season,  
439 meteorological change due to global warming will play a positive role on O<sub>3</sub> formation  
440 in most of China, with the enhancement within 0–4 ppb, but it will reduce the O<sub>3</sub> level  
441 in remote areas like Tibetan Plateau. The national average growth will be 0.3 ppb and  
442 that for YRD, PRD and BTH will be 1.9, 0.7, and 0.3 ppb, respectively. The response  
443 of O<sub>3</sub> to meteorological change is associated with some specific variables (Hong et al.,  
444 2019). For example, the great enhancement of O<sub>3</sub> in YRD might be attributable to a  
445 hotter, dryer and more stable atmosphere with growth of T-max (over 0.6 °C) and  
446 decline of RH and WS (Figure 2). The result is similar to Hong et al. (2019), which  
447 reported a change of 2–8 ppb of daily 1-hour maximum O<sub>3</sub> concentration for the peak  
448 season from the 2010s to 2050s under RCP4.5. In addition, the declining O<sub>3</sub> in Tibetan  
449 Plateau and the surrounding areas might result partly from the weakened long-range  
450 transport of peroxyacetyl nitrate (PAN, the principal NO<sub>x</sub> reservoir) from the polluted  
451 areas (Fischer et al., 2014). Driven by the elevated temperature, PAN from relatively  
452 polluted regions will undergo stronger thermal decomposition locally, thus fail to be  
453 transported far away to the remote regions to promote O<sub>3</sub> formation (Liu et al., 2013;  
454 Lu et al., 2019).

455 The influence of meteorological change on O<sub>3</sub> production is predicted to be much  
456 smaller for the non-warm season, with the magnitude within ±1 ppb in most areas and  
457 nationwide average at –0.2 ppb. In the three developed regions, the changes are  
458 predicted to range from –0.4 to 0.3 ppb, with little regional difference. The limited

459 influence might be attributable to the modest change in temperature and RH in the non-  
460 warm season.

### 461 **3.3.3 BVOCs and surrounding anthropogenic emission change**

462 Compared to domestic emissions, change of BVOCs emissions and anthropogenic  
463 emissions from surrounding countries will have a less influence (within  $\pm 3$  ppb) on  
464 surface O<sub>3</sub> concentration in China. BVOCs change tends to enhance O<sub>3</sub> while foreign  
465 emission change tends to restrain it in most areas (Figure 5).

466 The growing BVOCs emissions due to vegetation and climate change is estimated  
467 to enhance O<sub>3</sub> concentration by 0–3 ppb in the most areas of China, with a larger  
468 influence of 0.6 ppb in the warm season than that of 0.3 ppb in the non-warm season  
469 across the country (Figure 5c and 5g). In the warm season, relatively large growth of  
470 O<sub>3</sub> concentration will occur in BTH at 2.1 ppb, and those of YRD and PRD will be 1.5  
471 and 1.0 ppb, respectively. The abundant NO<sub>x</sub> emissions in BTH are expected to result  
472 in a larger O<sub>3</sub> concentration response to BVOCs emission change than YRD and PRD,  
473 even the BVOCs emission change of BTH will be smaller than the other two regions  
474 (Table 2). The result is in agreement with other numerical simulation experiments. Liu  
475 et al. (2019) reported a prominent O<sub>3</sub> enhancement even with a low BVOCs emission  
476 rate under RCP8.5, in a NO<sub>x</sub>-abundant environment. In the remote areas like Tibetan  
477 Plateau and part of Northeastern China, the increased BVOCs will remove O<sub>3</sub> due to  
478 the isoprene ozonolysis in low-NO<sub>x</sub> environment (Hollaway et al., 2017; Zhu et al.,  
479 2022). In general, regions with higher O<sub>3</sub> pollution levels and NO<sub>x</sub> emissions will suffer  
480 more risk of O<sub>3</sub> growing from rising BVOCs emissions in the future.

481 Most areas of China will benefit from the foreign emission change in terms of O<sub>3</sub>  
482 pollution alleviation (Figure 5d and 5h). An exception is Tibetan Plateau and its  
483 surrounding areas, which will be affected by the elevated emissions of NO<sub>x</sub> and VOCs  
484 from South Asia under SSP2-45. Limited by the range of pollutant transport, greater  
485 impacts will be found for coastal and border areas and less for inland areas (Ni et al.,  
486 2018). Larger O<sub>3</sub> changes in the three developed regions are predicted than that of the

487 whole country, benefitting from the precursor emission reduction in East Asia and  
488 Southeast Asia.

### 489 **3.4 The relationship between the joint and separate effects of multiple factors**

490 Figure 6 summarizes the contributions of individual factors to the total O<sub>3</sub> change  
491 by region and season. Due to the nonlinear response of O<sub>3</sub> to multi-factor changes, the  
492 aggregated contribution of the four factors does not equal to the joint contribution (i.e.,  
493 there exist gaps between the difference of the 2020s and 2060s and the aggregated  
494 contribution of four factors).

495 The varying domestic anthropogenic emissions are predicted to dominate the  
496 change of the future O<sub>3</sub>, with a relative contribution ranging from 75 % to 117 % for  
497 different regions and seasons. The relative contributions of the other three factors are  
498 estimated to be limited within  $\pm 25$  % at national and regional level. Among different  
499 regions, YRD will be more affected by climate change with the contribution of  $-13$  %  
500 and  $-12$  % for the warm and non-warm season, respectively, far greater than that of  
501 BTH and PRD ( $-6$  % to  $0$  %). BTH will be more affected by BVOCs emission change  
502 than other regions in the warm season ( $-21$  %), while YRD and PRD will be more  
503 affected in the non-warm season with the relative contributions of  $17$  % and  $-20$  %,  
504 respectively. Little regional difference is found for the relative contributions of foreign  
505 emission change.

506 To better understand the regional and seasonal differences of the relative  
507 contributions of future changes to O<sub>3</sub> concentration, we examine the nonlinear response  
508 of O<sub>3</sub> to precursor change in the three developed regions. We follow Chen et al. (2021)  
509 and Schroeder et al. (2017), and conduct a fit of lognormal distribution for the  
510 relationship of modeled hourly O<sub>3</sub> and NO<sub>2</sub> concentrations, as shown in Figure 7. The  
511 data points on the left of the turning point of fitted curve suggest a NO<sub>x</sub>-limited regime  
512 while on the right a VOC-limited regime, and data points around the turning point are  
513 under transitional regime.

514 The O<sub>3</sub>-NO<sub>2</sub> relationship from the 2020s to 2060s will be mostly influenced by the

515 changing domestic anthropogenic emissions, indicated by the close distributions of data  
516 points and fitted curves between “EMIS” and “2060s” in Figure 7. In the warm season,  
517 the future O<sub>3</sub>-NO<sub>2</sub> relations in BTH and YRD are predicted to change greatly from a  
518 highly O<sub>3</sub> polluted situation with moderate NO<sub>2</sub> concentration to a situation with a  
519 relatively low level of NO<sub>2</sub> (mostly under 10 ppb) and a moderate level of O<sub>3</sub> (under  
520 60 ppb). A weak VOC-limited regime appeared for the whole BTH in 2020s, consistent  
521 with recent observation-based analysis (Chen et al. 2023; Kong et al. 2024). There is  
522 big diversity within the region, including a dense area with strong VOC-limited regime  
523 and other areas with transitional or NO<sub>x</sub>-limited regime (Figure 7a). Represented by the  
524 moving of most points from the right of the turning point to near or left of the turning  
525 point, the NO<sub>x</sub>-limited and transitional regimes will dominate BTH in the 2060s.  
526 Compared to 2020s, the data points of 2060s are more closely distributed, indicating a  
527 reduced diversity of O<sub>3</sub> formation regime in the region. For YRD, most areas were  
528 under transitional or weak VOC-limited regime in the 2020s with limited diversity  
529 within the region, and the situation in 2060s will be similar to that of BTH (Figure 7a  
530 and 7b). The shift from weak VOC-limited regime in 2020s to transitional or NO<sub>x</sub>-  
531 limited regime in 2060s for BTH and YRD implies the influence of emission reduction  
532 on altering the sensitivity of O<sub>3</sub> formation to precursors. Most areas of PRD in the 2020s  
533 are under transitional or NO<sub>x</sub>-limited regimes, and the regime will transfer to a strong  
534 NO<sub>x</sub>-limited one in 2060s, with an almost positive correlation between NO<sub>2</sub> and O<sub>3</sub> in  
535 a low-NO<sub>2</sub> environment (Figure 7c). In the non-warm season, O<sub>3</sub> and NO<sub>2</sub> will remain  
536 negatively correlated for BTH and YRD till the 2060s, which suggests a persistent  
537 VOC-limited regime and explains the O<sub>3</sub> concentration growth along with substantial  
538 precursor emission reductions. The turning points are simulated at extremely low NO<sub>2</sub>  
539 concentrations of 2.0 and 1.2 ppb for BTH and YRD, respectively (Figure 7d and 7e).  
540 A big challenge still exists on effective emission controls to reduce the O<sub>3</sub> concentration  
541 in the non-warm season for the two regions. Differently, the O<sub>3</sub> formation sensitivity in  
542 most of PRD will shift from transitional regime towards a more NO<sub>x</sub>-limited situation

543 (Figure 7f). A simple comparison with the O<sub>3</sub> evolution and its precursor emission  
544 changes in developed country provided more policy implication. According to Chen et  
545 al. (2021) and the US Environmental Protection Agency, the northeastern US  
546 experienced rapid cross of the turning point of O<sub>3</sub> formation sensitivity during 1990s–  
547 2010s, with approximately 60 % reductions in both anthropogenic NO<sub>x</sub> and VOCs  
548 emissions. In BTH, the emissions are predicted to decline 57 % and 36 % for NO<sub>x</sub> and  
549 NMVOCs during 2020s–2060s under SSP2-45. Therefore, more ambitious reductions  
550 in NMVOCs will be necessary (ideally double the current projected abatement under  
551 SSP2-45), to accelerate the shift in the O<sub>3</sub> chemical regime for BTH.

552 The fitted curves of other three factors are similar to those of the 2020s, and the  
553 change of these factors will make little difference on NO<sub>2</sub> concentration but will result  
554 in moderate changes on O<sub>3</sub> concentration within  $\pm 2$  ppb. The limited changes of climate,  
555 BVOCs emissions and foreign anthropogenic emissions will not essentially alter the O<sub>3</sub>  
556 formation regime, but may change the O<sub>3</sub> production under the nearly same NO<sub>2</sub>  
557 concentration. Changes of individual meteorological factors are expected to easily  
558 influence the O<sub>3</sub> and NO<sub>2</sub> concentrations (Pope et al., 2015; Liu and Wang, 2020;  
559 Dewan and Lakhani, 2022). The modeled little response of NO<sub>2</sub> to meteorological  
560 change, except that in the non-warm season for BTH, might be attributed to the  
561 compensating effect of different variables. The limited influence of BVOCs on the O<sub>3</sub>  
562 formation sensitivity to precursors is consistent with Gao et al. (2022), which reported  
563 comparable empirical kinetic modelling approach (EKMA) curves with and without  
564 BVOCs emissions. The transboundary O<sub>3</sub> pollution results from the transport of both  
565 O<sub>3</sub> and its precursors (mainly associated with PAN), while NO<sub>2</sub> is less influenced by  
566 long-range transport due to its shorter lifetime (Ni et al., 2018; Yin et al., 2022).

567 The change in O<sub>3</sub> formation regime might partly explain the finding that the joint  
568 effect of multiple factors on restraining O<sub>3</sub> pollution will be larger than the aggregated  
569 effects of individual factors. Under a NO<sub>x</sub>-limited regime, O<sub>3</sub> is less sensitive to  
570 changing VOCs emissions (e.g., BVOCs emissions) than that under a VOC-limited one.

571 Therefore, the enhancement of O<sub>3</sub> due to BVOCs emission growth in the future will be  
572 restrained with a much lower NO<sub>2</sub> concentration. This indicates a co-benefit of reducing  
573 the anthropogenic emissions to restrain the potential O<sub>3</sub> pollution elevation due to  
574 growing BVOCs emissions (as a part of climate penalty) in the future.

### 575 **3.5 Change of O<sub>3</sub> exceedance events over the east of China**

576 Figure 8 shows the “O<sub>3</sub> exceedance events” over the east of China (mainly  
577 including Northern, Eastern, Central and Southern China) in the 2020s and 2060s, and  
578 the changes influenced by different factors. The exceedance is defined as number of  
579 days with the MDA8 O<sub>3</sub> exceeding the Chinese National Air Quality Standard-Grade  
580 II (160 μg m<sup>-3</sup> or 81.6 ppb). The exceedance events appear mainly in the warm season  
581 (Figure S7). Areas with frequent exceedance (over 50 days) in the 2020s were mainly  
582 located in Northern China. Much fewer exceedances are found for YRD and PRD (19.3  
583 and 8.2 days in 2020s, respectively). In the 2060s, the O<sub>3</sub> exceedance events will drop  
584 significantly. The exceedance days will be fewer than 10 days for most of the country,  
585 except for some areas in BTH which will still have more than 20 exceedance days over  
586 the year.

587 Domestic emission abatement will be the most important factor reducing the O<sub>3</sub>  
588 exceedance, particularly in Northern China. The exceedance days will be cut by 45.3,  
589 19.1 and 8.1 days for BTH, YRD and PRD, respectively, with the maximum reduction  
590 reaching 80 days within BTH and YRD. Notably, the spatial pattern of changing O<sub>3</sub>  
591 exceedance due to emission reduction is different from that of changing MDA8 O<sub>3</sub> due  
592 to emission reduction as shown in Figure 5a. Even the warm season MDA8 O<sub>3</sub>  
593 concentration of BTH will decline only 9.7 ppb, the O<sub>3</sub> exceedance events will be  
594 greatly reduced, indicating that national emission controls will be especially effective  
595 in reducing serious O<sub>3</sub> pollution. Climate change will mainly affect Jiangsu, Anhui,  
596 Henan and Hebei provinces, elevating the exceedance by more than 15 days in most of  
597 these areas. For YRD and PRD, climate change will elevate the exceedance by 9.5 and  
598 3.3 days, respectively. Some areas of BTH will benefit from climate change, with the

599 exceedance declining 0–10 days. The influences of BVOCs and foreign emission  
600 change on exceedance days are of limited regional differences, with a growth of 5 to 15  
601 days for the former and a decline of –5 to 0 days for the latter. The exceedances elevated  
602 by BVOCs emission growth will be 6.6, 6.1 and 2.8 days for BTH, YRD and PRD with  
603 the maximum reaching 19, 18 and 12 days within the region, respectively, reflecting an  
604 unneglectable role of biogenic source change on future O<sub>3</sub> episodes.

## 605 **4 Conclusions**

606 We explore the response of China's surface O<sub>3</sub> concentration to the future changes  
607 of multiple factors under SSP2-45, based on a series of sensitivity experiments with  
608 WRF-MEGAN-CMAQ simulations. From the 2020s to 2060s, the MDA8 O<sub>3</sub>  
609 concentration is predicted to decline by 7.7 and 1.1 ppb in the warm and non-warm  
610 season, respectively, and the O<sub>3</sub> exceedances of Chinese National Air Quality Standard  
611 (Grade II) will be largely eliminated. In the warm season, MDA8 O<sub>3</sub> in BTH, YRD and  
612 PRD will decline by 9.7, 14.8 and 12.5 ppb, respectively, larger than the national  
613 average level. However, MDA8 O<sub>3</sub> will increase in BTH and YRD in the non-warm  
614 season attributed to the reduced NO<sub>x</sub> emissions and thereby titration effect. The O<sub>3</sub>  
615 pollution will expand towards the non-warm season in the future, bringing new  
616 challenge for policy makers to optimize the strategy of precursor emission controls  
617 based on local conditions.

618 Reduction of local anthropogenic emissions is estimated to dominate the spatial  
619 distribution and magnitude of future O<sub>3</sub> change. Meteorological variation will lead to a  
620 change of MDA8 O<sub>3</sub> ranging between –1 and 4 ppb for most areas in the warm season.  
621 The influences of changing BVOCs and foreign anthropogenic emissions will be within  
622 ±3 ppb, with the former elevating O<sub>3</sub> while the latter reducing O<sub>3</sub>. Especially in areas  
623 with high O<sub>3</sub> pollution and intense NO<sub>x</sub> emissions, the growing BVOCs emissions will  
624 more enhance the risk of O<sub>3</sub> pollution. The joint effect of multiple factors on restraining  
625 O<sub>3</sub> pollution will be larger than the aggregated effects of individual factors, which can

626 be partly explained by the changing O<sub>3</sub> formation regime. Large amount of emission  
627 reduction under SSP2-45 will reshape the O<sub>3</sub> formation sensitivity to precursors. In  
628 BTH and YRD, O<sub>3</sub> formation in the warm season is projected to shift from weak VOC-  
629 limited to transitional or NO<sub>x</sub>-limited regime, while VOC-limited regime will still  
630 dominate in the non-warm season. In the future, O<sub>3</sub> will be less sensitive to BVOCs  
631 change in a low NO<sub>x</sub> environment along with persistent emission controls, highlighting  
632 the benefit of anthropogenic emissions abatement on mitigating the climate penalty and  
633 limiting O<sub>3</sub> pollution.

634 Limitations exist in current study. Firstly, the future climate data are taken from  
635 one single model CESM, subject to bias in the assessment of meteorological influence  
636 on O<sub>3</sub>. Secondly, some factors that will influence future O<sub>3</sub> level are not included in our  
637 analyses, such as the changing CH<sub>4</sub> concentration, increasing soil NO<sub>x</sub> emissions and  
638 the stratosphere-troposphere exchange of O<sub>3</sub>. For example, the hotspot of soil NO<sub>x</sub>  
639 emissions in northern China is also the region with large reduction of anthropogenic  
640 NO<sub>x</sub> emission but relatively small decline in O<sub>3</sub> concentrations. Under a warmer climate,  
641 a growing trend of soil NO<sub>x</sub> emissions is expected for the future, and may thus present  
642 an additional challenge for O<sub>3</sub> pollution alleviation. Thirdly, there exist gaps between  
643 the downscaled and realistic conditions of meteorology for the 2020s, leading to  
644 uncertainty in the O<sub>3</sub> simulation. Finally, the changing O<sub>3</sub> formation regime is presented  
645 through the relation between O<sub>3</sub> and NO<sub>2</sub> concentrations, and the mechanism how the  
646 climate penalty will influence O<sub>3</sub> formation under substantial reduction of  
647 anthropogenic emissions needs to be better analyzed in future studies.

## 648 **Data availability**

649 All data in this study are available from the authors upon request.

## 650 **Author contributions**

651 JYang developed the methodology, conducted the work and wrote the draft. YZhao  
652 improved the methodology, supervised the work and revised the manuscript. YWang



653 and LZhang contributed to the methodology and provided supports to the scientific  
654 interpretation and discussions.

### 655 **Competing interests**

656 The authors declare that they have no conflict of interest.

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## 1012 **FIGURE CAPTIONS**

1013 Figure 1 The modelling domain and geographical definitions (denoted by colors) of this  
1014 study. Boundaries of the three regions, including BTH (Beijing-Tianjin-Hebei), YRD  
1015 (Yangtze River Delta) and PRD (Pearl River Delta), are marked by dark grey lines.

1016 Figure 2 Projected changes of the ozone-related meteorological factors, including daily  
1017 maximum temperature at 2 m (T-max, a and d), relative humidity (RH, b and e) and  
1018 wind speed (WS, c and f), from the 2020s to 2060s. Panels (a-c) represent those of the  
1019 warm season, and panels (d-f) represent those of non-warm season.

1020 Figure 3 Simulation and projection of seasonal average MDA8 O<sub>3</sub> in the 2020s (Case1,  
1021 a and b) and 2060s (Case2, d and e), and the changes over this period (Case2–Case1, c  
1022 and f). Panels (a-c) represent those of the warm season, and panels (d-f) represent those  
1023 of non-warm season. Regional mean concentrations across China (CHN), BTH, YRD  
1024 and PRD are inset.

1025 Figure 4 Simulation and projection of monthly average MDA8 O<sub>3</sub> in the 2020s and  
1026 2060s across CHN (a), BTH (b), YRD (c) and PRD (d).

1027 Figure 5 Projected changes of MDA8 O<sub>3</sub> from the 2020s to 2060s attributed to  
1028 anthropogenic emissions from local sources (Case3–Case1, a and e), meteorological  
1029 conditions (Case4–Case1, b and f), BVOCs emissions (Case5–Case1, c and g) and  
1030 anthropogenic emissions from surrounding countries (Case6–Case1, d and h). Panels  
1031 (a-d) represent those of the warm season, and panels (e-h) represent those of non-warm  
1032 season. Regional mean changes across CHN, BTH, YRD and PRD are inset.

1033 Figure 6 The relationships between the separate MDA8 O<sub>3</sub> changes attributed to the  
1034 four factors (denoted by the name of Case3–6) and the total changes from the 2020s to  
1035 2060s over China and the three regions. Panels (a-d) represent those of the warm season,  
1036 and panels (e-h) represent those of non-warm season. The relative contributions of the



1037 four factors to the total influence of future change are shown in the light grey box.

1038 Figure 7 The relationships between simulated hourly NO<sub>2</sub> and O<sub>3</sub> concentrations with  
1039 the lognormal fits for different regions and seasons. The colored circles, representing  
1040 different cases, come from the seasonal average concentrations for each grid in the  
1041 target region. The specific circles with black border represent the regional average  
1042 situation, and the turning points of every fitted curve are marked by the “+” sign. The  
1043 density plots of the 2020s and 2060s are inset.

1044 Figure 8 Projected annual O<sub>3</sub> exceedance over the east of China in the 2020s and 2060s,  
1045 and the exceedance changes when the four factors at 2060s level. Regional mean  
1046 changes across CHN, BTH, YRD and PRD are inset.

1047

1048 **TABLES**

1049 **Table 1 List of simulation cases to investigate the impact of future change upon**  
 1050 **surface O<sub>3</sub> in China, with sensitivity experiments from the perspectives of four**  
 1051 **main influencing factors.**

Case number	Case name	China's local emissions	Meteorological conditions	BVOCs emissions	Surrounding emissions
Case1	2020s	2020	2018-2022	2018-2022	2020
Case2	2060s	2060	2058-2062	2058-2062	2060
Case3	EMIS	2060	2018-2022	2018-2022	2020
Case4	CLIM	2020	2058-2062	2018-2022	2020
Case5	BVOC	2020	2018-2022	2058-2062	2020
Case6	SURR	2020	2018-2022	2018-2022	2060

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1054 **Table 2 The BVOCs estimation over China and emission intensity in BTH, YRD**  
 1055 **and PRD of the 2020s and 2060s, as well as the corresponding growth rates over**  
 1056 **this period.**

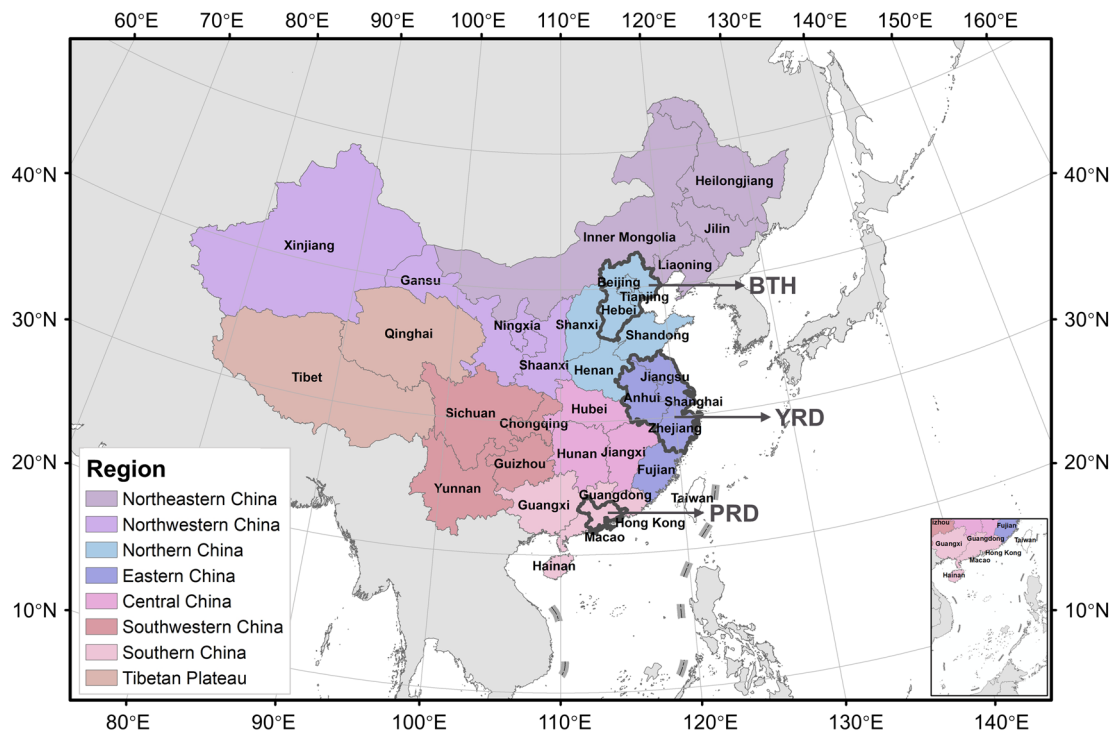
	China	BTH	YRD	PRD
	Emissions (Tg)	Emission intensity (Gg grid <sup>-1</sup> )		
2020s	33.6	1.4	4.6	8.7
2060s	43.4	1.7	5.7	10.7
Growth rate	29.2 %	21.4 %	23.9 %	23.0 %

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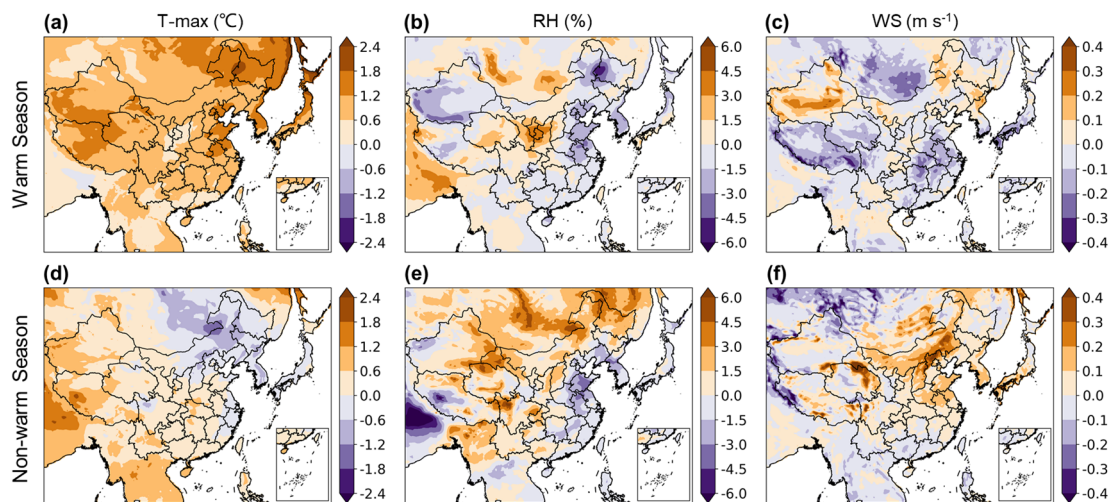
1059 **FIGURES**

1060 **Figure 1**

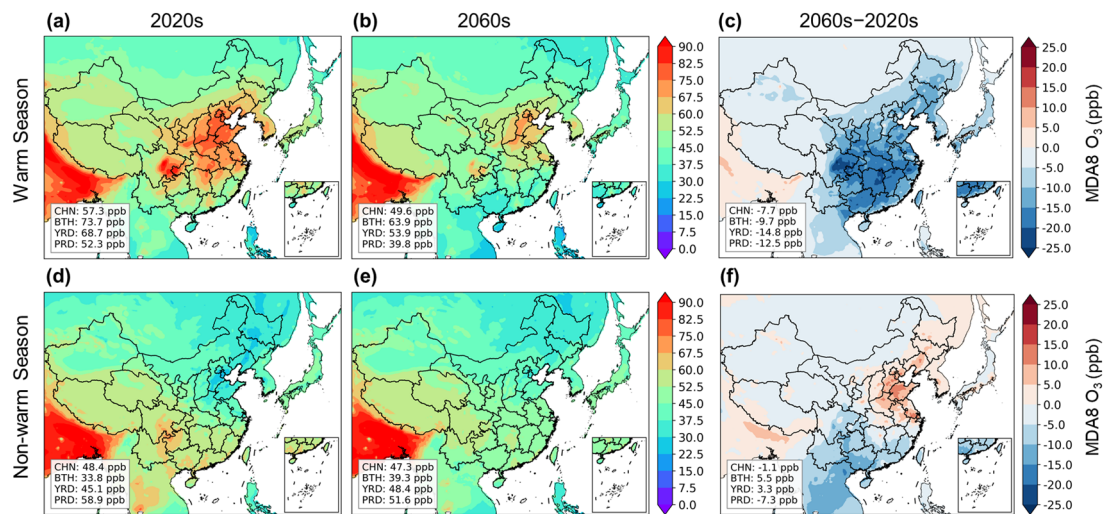


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1063 **Figure 2**



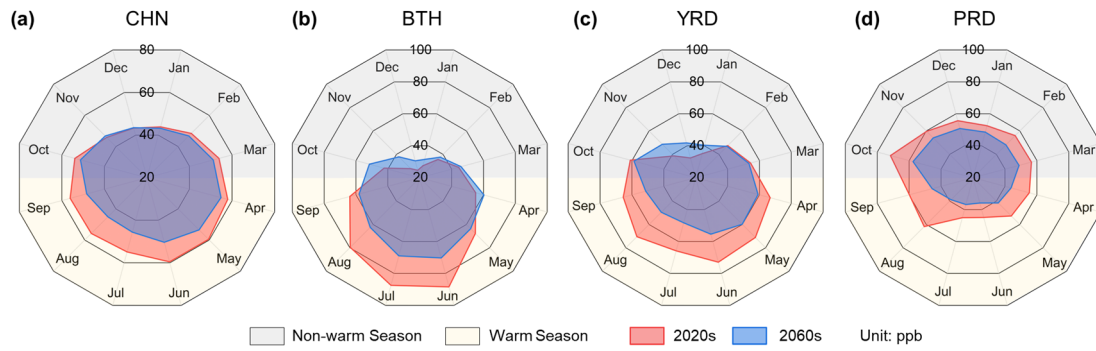
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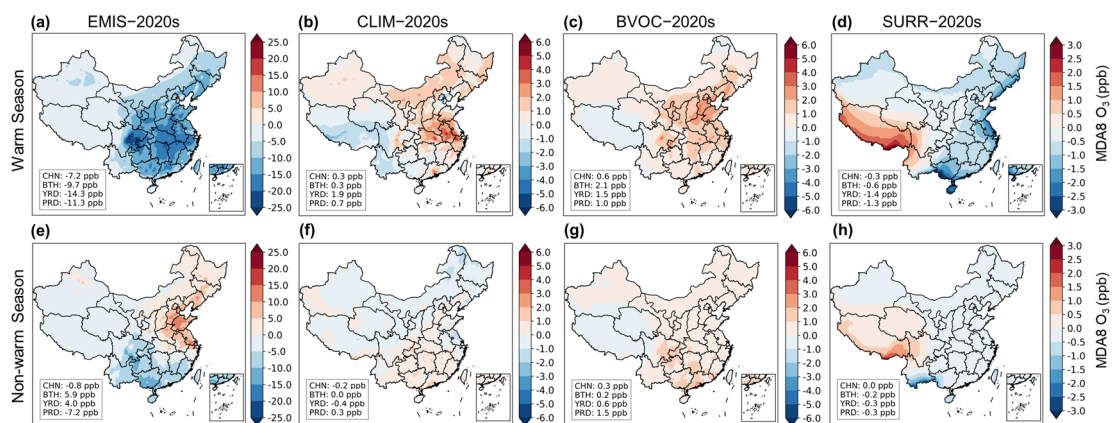
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1069 **Figure 4**



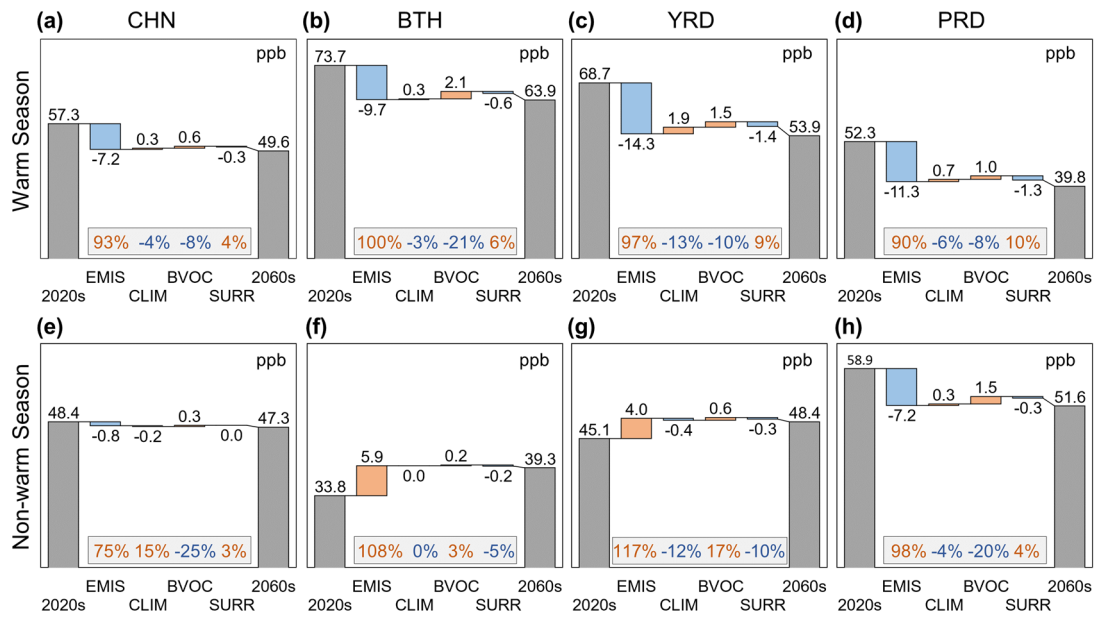
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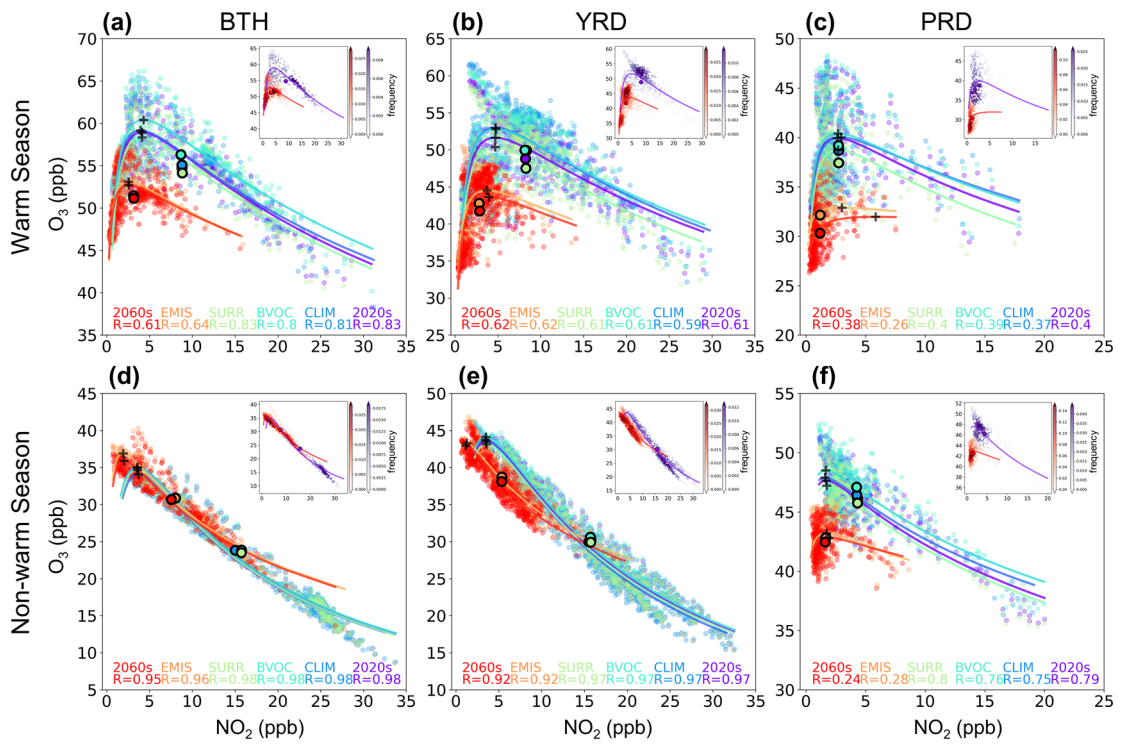
1075 **Figure 6**



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1081 **Figure 8**

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