

1 **Meteorological characteristics of severe ozone pollution events in**
2 **China and their future predictions**

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23 Abstract

24 Ozone (O₃) has become one of the most concerning air pollutants in China in recent
25 decades. In this study, based on surface observations, reanalysis data, ~~and~~ global atmospheric
26 chemistry model simulations and multi-model future predictions, meteorological
27 characteristics conducive to severe O₃ pollution in various regions of China are investigated,
28 and their historical changes and future trends are analyzed. During the most severe O₃ ~~pollution~~
29 polluted months ~~over the North China Plain (NCP) and Yangtze River Delta (YRD)~~, the
30 chemical production of O₃ is enhanced under the hot and dry conditions over the North China
31 Plain (NCP) in June 2018 and Yangtze River Delta (YRD) in July 2017, while the regional
32 transport is the main reason causing the severe O₃ pollution over Sichuan Basin (SCB) July
33 2015 and Pearl River Delta (PRD) in September 2019 ~~during the severe polluted months~~. Over
34 the last four decades, the frequencies of high temperature and low relative humidity conditions
35 increased in 2000-2019 relative to 1980-1999, indicating that O₃ pollution in both NCP and
36 YRD became more frequent under the historical climate change. In SCB and PRD, the
37 occurrence of atmospheric circulation patterns similar to those during the polluted months
38 increased, together with the more frequent hot and dry conditions, contributing to the increases
39 in severe O₃ pollution in SCB and PRD during 1980–2019. In the future (by 2100), the
40 frequencies of months with anomalous high temperature show stronger increasing trends in the
41 high forcing scenario (SSP5-8.5) compared to the sustainable scenario (SSP1-2.6) in China. It
42 suggests that high anthropogenic forcing will not only lead to slow economic growth and
43 climate warming, but also likely result in environmental pollution issues.

44 **1. Introduction**

45 Tropospheric ozone (O₃), one major air pollutant, is formed in photochemical reactions of
46 nitrogen oxides (NO_x) and volatile organic compounds (VOCs) when exposed to sunlight
47 (Finlayson-Pitts and Pitts, 1997; Silman, 1999). Enhanced O₃ pollution harms ecosystems and
48 human health (Fleming et al., 2018; Maji et al., 2019) by reducing crop yields (Ainsworth et
49 al., 2012; Mills et al., 2018) and aggravating cardiopulmonary disease (Ebi and McGregor,
50 2008; Liu et al. 2018). In recent years, near-surface ozone concentrations in many regions of
51 China have been increasing considerably (Verstraeten et al., 2015; Cheng et al., 2019; Zhang
52 et al., 2020, Li et al., 2019; Lu et al., 2018; Silver et al., 2018; Yin et al., 2019, Lu et al., 2020).
53 Lu et al. (2020) revealed that the daily maximum of 8-h average O₃ concentration (MDA8-O₃)
54 in China increased by 2.4 ppb per year (5.0% relative to the average) during April–September
55 over 2013–2019.

56 In addition to emissions, O₃ concentrations are influenced by meteorological factors such
57 as temperature, relative humidity, solar radiation, and winds (Mott et al., 2005; Fu and Tian,
58 2019; Gong and Liao, 2019; Li et al., 2019, 2020; Le et al., 2020; Zhao et al., 2020). Typically,
59 strong solar radiation, high surface air temperatures, and low relative humidity are conducive
60 to photochemical production of O₃, causing a raise of O₃ concentration (Peterson and Flowers,
61 1977; Xu, et al., 2011; Coates et al., 2016; Li et al., 2020; Dang et al., 2021). Wind speed is
62 negatively correlated with surface O₃ because low wind speed facilitates the accumulation of
63 O₃ upon production (Zhang et al., 2015; Wang et al., 2017; Liu and Wang, 2020). Han et al.
64 (2020) explored the impacts of various meteorological factors on the daily variation of summer
65 surface O₃ in eastern China based on a multiple linear regression method and suggested that

66 relative humidity is the primary factor affecting O₃ concentration in central and south parts of
67 eastern China, while temperature is the most important factor governing O₃ concentration in
68 north of eastern China. Gong and Liao (2019) reported that the meteorological characteristics
69 of O₃ pollution events in North China during 2014–2017 were the high daily maximum
70 temperature, low relative humidity, abnormal southerly winds and high pressure at 500 hPa.
71 These findings emphasize that meteorological factors play a crucial role in regulating O₃
72 pollution in China.

73 Atmospheric circulation patterns affect O₃ concentrations over China through changing
74 meteorological factors (Yang et al, 2014, [2022](#); Zhao and Wang, 2017; Shu et al., 2019; Dong
75 et al., 2020; Zhou et al., 2022). Zhao and Wang (2017) examined the influence of the Western
76 Pacific Subtropical high (WPSH) on O₃ over eastern China based on observations and
77 reanalysis data from 2014 to 2016. They found that stronger WPSH enhanced the moisture
78 transport to southern China, which was detrimental to the photochemical reaction of O₃,
79 leading to a decrease in surface O₃ concentration in southern China, whereas O₃ concentrations
80 in northern China increased under the stronger WPSH related to the dry and hot conditions
81 favoring O₃ production. On the basis of observational O₃ data and ERA5 reanalysis data during
82 2014–2018, Dong et al. (2019) analyzed the impact of synoptic patterns on summertime O₃
83 pollution in the North China Plain and revealed that the most severe O₃ pollution weather
84 pattern is associated with anomalous southwesterly winds, which carry dry, warm air from
85 inland southern China to the North China Plain and favor the chemical production of O₃. Zhou
86 et al. (2022) explored the impacts of Asian summer monsoon on the interannual variation of
87 O₃ concentrations based on surface measurements and GEOS-Chem model simulations. They

88 showed that the East Asian summer monsoon strength was positively correlated with O₃
89 concentration in south-central China and South Asian summer monsoon has complex effects
90 on O₃ pollution in China, mainly through changing transboundary transport related to large-
91 scale circulations.

92 As mentioned above, many previous studies have examined the meteorological
93 characteristics of O₃ pollution in China in limited regions in China. However, they focused on
94 O₃ pollution over limited regions in China in each study (e.g., the North China Plain, southern
95 China). These studies only examined the meteorological characteristics— of O₃ pollution in a
96 short time period due to the lack of observational data and did not consider the historical and
97 future trends of these meteorological factors. In this study, the meteorological characteristics
98 conducive to severe O₃ pollution in several polluted areas of China, including the North China
99 Plain (NCP), Yangtze River Delta (YRD), Sichuan Basin (SCB), and Pearl River Delta (PRD),
100 are respectively investigated based on the observed surface O₃ concentrations, reanalysis data,
101 and GEOS-Chem model simulations. Besides, the contributions from various chemical and
102 physical processes inducing regional O₃ pollution are quantified using an integrated process
103 rate (IPR) analysis method. The historical changes in these meteorological factors favoring
104 severe O₃ pollution over 1980–2019 are provided. Moreover, variations ~~of~~ in future
105 meteorological patterns during 2021–2100 leading to severe O₃ pollution in China are
106 presented under the sustainable and high forcing scenarios according to the multi-model data
107 from the Coupled Model Intercomparison Project Phase 6 (CMIP6).

108 **2. Methods**

109 **2.1 Surface ozone observations and meteorological reanalysis**

110 Hourly surface O₃ concentrations are obtained from the Ministry of Ecology and
111 Environment (MEE) of China. The observational network was established in 2013 with 450
112 monitoring sites and increased to 1,500 monitoring sites by 2019, covering about 360 cities in
113 China. MDA8-O₃ are calculated based on hourly O₃ concentrations from April-September
114 during 2013 to 2020. In this study, O₃ pollution days are defined as the days when MDA8-O₃
115 exceeds 160 µg m⁻³ according to the China National Ambient Air Quality Standard (GB3095-
116 2012).

117 The meteorological fields are taken from the European Centre for Medium-Range
118 Weather Forecasts (ECMWF) ERA5 monthly reanalysis dataset during 1980–2020, with a
119 horizontal resolution of 0.25° × 0.25°. To explore the meteorological characteristics that are
120 conducive to O₃ pollution, sea level pressure (SLP), geopotential height (GPH) at 500 hPa,
121 wind fields at 850 hPa and 500 hPa, temperature at 2m (T2m) and surface relative humidity
122 (RH) are adopted, which can have significant impacts on O₃ variations in China (Jiang et al.,
123 2020; Dong et al., 2020; Le et al., 2020).

124 **2.2 GEOS-Chem model simulations**

125 O₃ concentrations and the related chemical and physical processes causing O₃ variations
126 over 1981–2020 are simulated in the global atmospheric chemistry model GEOS-Chem
127 (version V12.9.3), driven by the Modern-Era Retrospective analysis for Research and
128 Application, Version 2 (MERRA-2). Simulations are performed on 47 vertical layers from
129 surface to 0.01 hPa, and a horizontal grid of 2° latitude × 2.5° longitude. GEOS-Chem model
130 incorporates a fully coupled O₃-NO_x-hydrocarbon-aerosol chemical mechanism (Pye et al.,
131 2009; Mao et al., 2013; Sherwen et al., 2016). Boundary-layer mixing uses a non-local scheme

132 (Lin and McElroy, 2010), and stratospheric O₃ chemistry employs the linearized O₃
133 parameterization (LINOZ) (McLinden et al., 2000).

134 Global anthropogenic aerosol and precursor gas emissions driving the simulations are
135 from the Community Emissions Data System (CEDS, Hoesly et al., 2018) and biomass burning
136 emissions are from the Global Fire Emissions Database, Edition 4 (GFED4, Van der Werf et
137 al., 2017). VOCs emissions from biogenic sources are provided offline by the Model of
138 Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN V2.1, Guenther et al.,
139 2012). Lightning and soil emissions are specified in the model (Hudman et al., 2012; Ott et al.,
140 2010). Anthropogenic emissions in China are updated with the Multi-resolution Emission
141 Inventory (MEIC), a localized emission dataset for China. Anthropogenic, biomass burning,
142 biological and other natural emissions are kept at 2017 levels during the simulations, so as to
143 eliminate the influence of emission changes on the interannual variation and trends of O₃.
144 Simulated O₃ distributions with the same configuration in GEOS-Chem have been extensively
145 evaluated in many studies, and the model has been reported to capture O₃ concentrations well
146 in China (e.g., Li et al., 2019; Lu et al., 2019; Ni et al., 2018).

147 **2.3 CMIP6 multi-model simulations**

148 The multi-model simulations from historical and the Scenario Model Intercomparison
149 Project (ScenarioMIP) in CMIP6 are used to analyze the historical variations and future trends
150 of meteorological conditions conducive to severe O₃ pollution. Two different future scenarios
151 of the Shared Socioeconomic Path (SSPs) are applied, including the sustainable scenario
152 (SSP1-2.6) and the high forcing scenario (SSP5-8.5). Totally simulations from 13 models
153 (ACCESS-CM2, ACCESS-ESM1-5, CAS-ESM2-0, CMCC-CM2-SR5, CMCC-ESM2,

154 FGOALS-f3-L, FGOALS-g3, GFDL-ESM4, INM-CM4-8, INM-CM5-0, IPSL-CM6A-LR,
155 MPI-ESM1-2-HR, MPI-ESM1-2-LR) are analyzed in this study.

156 **3. Results**

157 **3.1 Meteorological characteristics conducive to regional ozone pollution**

158 To investigate the relationship between meteorological conditions and regional O₃
159 pollution in China, the frequencies of O₃ pollution days from April to ~~September~~October
160 during 2013–2020 are calculated for Beijing, Shanghai, Chengdu and Guangzhou, representing
161 the typical four polluted regions in China (i.e., NCP, YRD, SCB and PRD) (Figure 1).
162 Observational data show the highest frequencies of O₃ pollution days in June 2018, July 2017
163 and September 2019 in Beijing, Shanghai and Guangzhou, with pollution days up to 22, 20 and
164 19 days per month, respectively. The top three highest frequencies of O₃ pollution days in
165 Chengdu are in July 2016, July 2015 and July 2018 (16, 15 and 15 days per month,
166 respectively). Variations in O₃ concentration in the real world are driven by changes in both
167 meteorological factors and emissions. With fixed emissions, the positive anomalies of near-
168 surface O₃ concentrations over NCP, YRD and PRD during their most polluted months can also
169 be reproduced by the GEOS-Chem model (Figure 2), suggesting that the O₃ pollutions during
170 the most polluted months over NCP, YRD and PRD are likely attributable to the anomalies of
171 meteorological conditions. In the top three O₃ polluted months in Chengdu, only in July 2015
172 the higher concentrations than the long-term averages can be captured by the
173 ~~simulation~~simulations with fixed emissions. Therefore, in this study, we focus on the
174 meteorological characteristics in June 2018, July 2017, July 2015 and September 2019, that
175 were conducive to the severe O₃ pollution over NCP, YRD, SCB and PRD, respectively.

176 When O₃ pollution was the most severe over NCP in June 2018, an anomalous high
177 pressure occurred at 500 hPa over NCP (Fig. 3b), relative to the 40-year climatological
178 averages from 1980 to 2019, leading to positive T2m anomalies near the surface (Fig. 3c).
179 Anomalous lows located over northeastern China and northwestern Pacific (Fig. 3a) and the
180 associated anomalous northerly winds prevent the moisture moving from the ocean to NCP,
181 causing negative RH anomalies over NCP (Fig. 3d). The meteorological conditions with the
182 high T2m and low RH are favorable for the photochemical production of O₃. When the most
183 severe O₃ pollution occurred in July 2017, YRD was dominated by anomalous high pressure
184 in the lower and middle troposphere (Figs. 4a and 4b). Under the control of high pressure, the
185 meteorological conditions (e.g., high T2m and low RH) enhance the photochemical production
186 of O₃ (Figs. 4c and 4d). In the O₃ pollution event of SCB in July 2015, the negative T2m
187 anomaly is not conducive to the O₃ production (Fig. 5c), although the RH was low (Fig. 5d).
188 Meanwhile, the anomalous low over eastern China and northwestern Pacific in the middle
189 troposphere favors regional O₃ transport from the polluted source region over eastern China to
190 SCB (Fig. 5b) and the anomalous high over central-western China is conducive to the vertical
191 transport of upper tropospheric O₃ down to the lower troposphere (Fig. 5a). For the PRD in
192 September 2019, the anomalous high covering almost the entire China along with the
193 anomalous low over East China Sea generates northerly wind anomalies in the lower
194 troposphere over eastern China, which tend to transport polluted air from northern China and
195 weaken the inflow of oceanic clean air (Fig. 6). The temperature increase is much more
196 significant in the upwind regions as compared to PRD, suggesting that the strong regional
197 transport could be the primary reason causing this severe O₃ pollution event of PRD.

198 **3.2 Physical and chemical mechanisms leading to regional ozone pollution**

199 To further explore the mechanisms of meteorological changes leading to the severe O₃
200 pollution over the four typical polluted regions in China, contributions of individual chemical
201 and physical processes to O₃ variations are quantified based on the IPR analysis from GEOS-
202 Chem simulations and summarized in Table 1.

203 Consistent with the meteorological anomalies analyzed above, high temperature and low
204 RH meteorological conditions in NCP are conducive to the photochemical production of O₃.
205 During the polluted month over NCP, the chemical production of tropospheric O₃ is higher than
206 the long-term average by 2.36 Gg day⁻¹, while the horizontal transport also contributes to the
207 increase in O₃ mass by 1.58 Gg day⁻¹ (Table 1). Due to the enhanced northwesterly winds, the
208 import of O₃ mass from the north and east-west of NCP was increased by 1.80 and 0.62 Tg,
209 respectively (Table 2). In YRD, the chemical production (2.38 Gg day⁻¹) is also the dominant
210 process that drives the O₃ concentration increase during the severe polluted month, associated
211 with the warm and dry conditions. Therefore, the anomalous chemical production is the major
212 process that induced O₃ pollution in NCP and YRD during the severe polluted months.

213 Different from NCP and YRD, horizontal transport is the main process that caused O₃
214 pollution in SCB and PRD during the severe polluted months. It contributes to the rate of
215 increase in O₃ mass by 5.10 and 6.67 Gg day⁻¹, respectively, over SCB and PRD, while other
216 processes tend to decrease the O₃ mass (Table 1). Due to the anomalous northerly winds over
217 SCB, more O₃ is transported into SCB from north (by 4.02 Tg), and the anomalous
218 northeasterly winds enhance the O₃ transport from the north and east of PRD by 1.97 and 1.09
219 Tg, respectively, leading to the increase in O₃ concentrations over SCB and PRD during the

220 severe polluted months relative to the climatological averages (Table 2). Note that, the chemical
221 production of tropospheric O₃ decreased in SCB and PRD during the severe polluted months.
222 It could have been biased by the relatively coarse model resolution in this study (2° latitude
223 × 2.5° longitude), since that the SCB and PRD for calculating the chemical and physical
224 processes only cover limited grid boxes. Further studies should be performed using a model
225 with finer resolution or a nested simulation method.

226 **3.3 Historical and future changes in the meteorological conditions**

227 O₃ pollution has deteriorated in China during recent decades, which could be related to
228 the changes in meteorological conditions. Time series of T2m and RH anomalies in the polluted
229 months during the 1980–2019 and frequencies of high T2m and low RH months during 1980–
230 1999 and 2000–2019 over the four polluted regions in China based on ERA5 reanalysis data
231 are shown in Figure 7. Due to climate change, both the high temperature and low RH conditions
232 in NCP, YRD, SCB and PRD all increased during the past four decades (2000-2019 versus
233 1980-1999). Based on the analysis showing that chemical production is the dominant process
234 of severe O₃ pollution in NCP and YRD, the increases in the frequency of high temperature
235 and low RH indicate that severe O₃ pollution in both NCP and YRD has become more frequent
236 under the historical climate change. In SCB and PRD, the severe O₃ pollution is more related
237 to changes in regional transport. Similar to the analyzing method used in previous studies (Li
238 et al., 2018; Yang et al., 2021), tThe SLP and 500 hPa GPH over East Asia and Western Pacific
239 in the same month of each year –similar to those during the severe polluted months in both
240 SCB and PRD have increased (2000-2019 versus 1980-1999) (Figure 8), together with the
241 more frequent hot and dry conditions (Figure 7), leading to the increases in severe O₃ pollution

242 in SCB and PRD during 1980–2019.

243 Many studies have reported that future climate change will have significant influences
244 on O₃ pollution in China through changing meteorological factors (e.g., Li et al., 2023; Wang
245 et al., 2022). Here, the frequencies of extreme months with high T2m and low RH and the
246 frequencies of extreme months with SLP and 500 hPa GPH that have moderate to high
247 correlation to those in the polluted months in the four regions of China, under the sustainable
248 (SSP1-2.6) and high forcing (SSP5-8.5) scenarios during 2021–2100 from CMIP6 multi-model
249 results, are presented in Figures 9 and 10, respectively. Unlike the historical changes in the
250 meteorological conditions that caused the severe O₃ pollution through chemical production and
251 regional transport, future variations in meteorological conditions conducive to the severe O₃
252 pollution are more related to the global warming process that enhances the O₃ production in
253 China. The frequencies of months with anomalous high temperature show obvious upward
254 trends in both SSP1-2.6 and SSP5-8.5 scenarios over the four regions, and the increasing trends
255 in SSP5-8.5 are much more significant than in SSP1-2.6. Frequencies of low RH months show
256 downward trends in NCP, YRD and SCB, especially under SSP5-8.5, while there is an upward
257 trend in PRD. Note that the trends in frequencies of low RH months are much less significant
258 than in high temperature months. The frequencies of extreme months with SLP and 500 hPa
259 GPH that are similar to those in the severe O₃ pollution months in the four regions do not show
260 significant trends in the SSPs. Hence, the future climate change may aggregate O₃ pollution in
261 China by enhancing the chemical production related to temperature increases. The O₃ pollution
262 exacerbation is projected to be less significant in the sustainable scenario due to the moderate
263 temperature increase than in the high forcing scenario, suggesting that the sustainable scenario

264 is the optimal path to retaining clean air in China. High anthropogenic radiative forcing will
265 not only lead to slow economic growth and climate warming, but also result in the
266 environmental pollution.

267 **4. Conclusions and Discussions**

268 O₃ pollution harms ecosystems and human health. In recent years, near-surface O₃
269 concentrations in many regions of China have been increasing considerably. Base on
270 observational O₃ data, ERA5 reanalysis data and GEOS-Chem model simulations,
271 meteorological characteristics conducive to severe O₃ pollution in different regions of China
272 are investigated in this study. Contributions from various chemical and physical processes
273 inducing O₃ pollution are quantified using an integrated process rate (IPR) analysis method.
274 Furthermore, historical changes and future trends of meteorological conditions leading to
275 severe O₃ pollution in China are explored based on the meteorological reanalysis and CMIP6
276 multi-model future predictions, which is of great implication for the mitigation and prevention
277 of O₃ pollution over China.

278 In this study, June 2018, July 2017, July 2015 and September 2019 are identified as the
279 most severe O₃ pollution months influenced by meteorological factors over NCP, YRD, SCB
280 and PRD, respectively. Severe O₃ pollution in June 2018 over NCP and in July 2017 over YRD
281 is mainly due to enhanced chemical production related to hot and dry conditions. The chemical
282 production of O₃ in the severe polluted months over NCP and YRD are 2.36 Gg day⁻¹ and 2.38
283 Gg day⁻¹, respectively, higher than the climatological averages. Different from NCP and YRD,
284 regional transport is the main process leading to the high O₃ concentration in SCB and PRD
285 during the respective severely polluted months, which contributes to the rate of increase in O₃

286 mass by 5.10 and 6.67 Gg day⁻¹, respectively, over SCB and PRD. During the severely polluted
287 months, related to large-scale circulation patterns, anomalous northerly winds transport more
288 O₃ into SCB from north, and anomalous northeasterly winds enhance the O₃ transport from the
289 north and east into PRD.

290 Over the last four decades (2000-2019 versus 1980-1999), the frequencies of high
291 temperature and low RH increased, indicating that O₃ pollution in both NCP and YRD has
292 become more frequent under the historical climate change. In SCB and PRD, the occurrence
293 of atmospheric circulation patterns similar to those during the polluted months in both SCB
294 and PRD has increased, together with the more frequent hot and dry conditions, leading to the
295 increases in severe O₃ pollution in SCB and PRD during 1980–2019. In the future (by 2100),
296 the frequencies of months with anomalous high temperature show obvious upward trends in
297 both sustainable (SSP1-2.6) and high forcing (SSP5-8.5) scenarios over the four regions, and
298 the increasing trends in SSP5-8.5 are much more significant than in SSP1-2.6. This suggests
299 that high anthropogenic radiative forcing will not only lead to slow economic growth and
300 climate warming, but also likely result in environmental pollution issues. The sustainable
301 scenario is the optimal path to retaining clean air in China.

302 There are some limitations and uncertainties in this work that can be further addressed in
303 future studies. For example, the model only captures the high O₃ concentrations in July 2015
304 in Chengdu among its top three polluted months. It is probably because the emissions are kept
305 at 2017 levels during the simulations. The high O₃ anomalies in July 2016 and July 2018 are
306 more likely influenced by the interannual changes in local precursor emissions in the
307 background of country-level increases in O₃ concentration in recent years. However, we also

308 can not rule out the possible inaccuracy in the model simulations to interpret severe O₃
309 pollution events in the SCB, which deserves further investigation with multi-model simulations.
310 In addition, this study focuses on the most extreme O₃ pollution in several polluted areas of
311 China. However, many other meteorological conditions can also cause O₃ pollution, although
312 they may not be as extreme as the cases analyzed in this study, which requires comprehensive
313 analysis for individual regions in future studies. Although the historical changes in the
314 meteorological patterns causing severe O₃ pollution are in accordance with the elevated O₃
315 levels in China in the recent decade, the quantitative analysis of meteorological impacts needs
316 full consideration of factors leading to O₃ pollution, including changes in anthropogenic and
317 natural emissions of its precursors, O₃ chemical regime, other meteorological factors conducive
318 to O₃ pollution, and stratosphere-troposphere exchange.

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563 **Code and data availability.** The GEOS-Chem model is available at
564 <https://zenodo.org/record/3974569#.YTD81NMzagR> (last access: June 2023). O₃ observations
565 over China can be obtained at <https://quotsoft.net/air> (last access: June 2023). ERA5 reanalysis
566 data can be downloaded at [https://www.ecmwf.int/en/forecasts/datasets/reanalysis-](https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5)
567 [datasets/era5](https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5) (last access: June 2023). The multi-model simulations of the Coupled Model
568 Intercomparison Project Phase 6 (CMIP6) are from <https://esgf-node.llnl.gov/search/cmip6/>
569 (last access: June 2023).

570 **Author contribution.** YY designed the research; YY and YZ performed simulations and
571 analyzed the data. All authors including HW, LH, PW, and HL discussed the results and wrote
572 the paper.

573 **Competing interests.** At least one of the (co-)authors is a member of the editorial board of
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584 **Table 1.** Anomalies in net rate of changes in tropospheric O₃ mass (Gg day⁻¹) over NCP (115°–
585 120°E, 38°–44°N), YRD (120°–125°E, 28°–32°N), SCB (102.5°–105°E, 30°–32°N) and PRD
586 (110°–115°E, 22°–26°N) due to physical and chemical processes in the most polluted months
587 (June 2018, July 2017, July 2015 and September 2019, respectively) relative to the same
588 months averaged during 1981–2019.

589

| | Beijing | Shanghai | Chengdu | Guangzhou |
|------------------------------|---------|----------|---------|-----------|
| Chemical reaction | 2.36 | 2.38 | -2.80 | -1.52 |
| Horizontal transport | 1.58 | -1.18 | 5.10 | 6.67 |
| Diffusion and dry deposition | 0.29 | 0.24 | -0.73 | -0.93 |

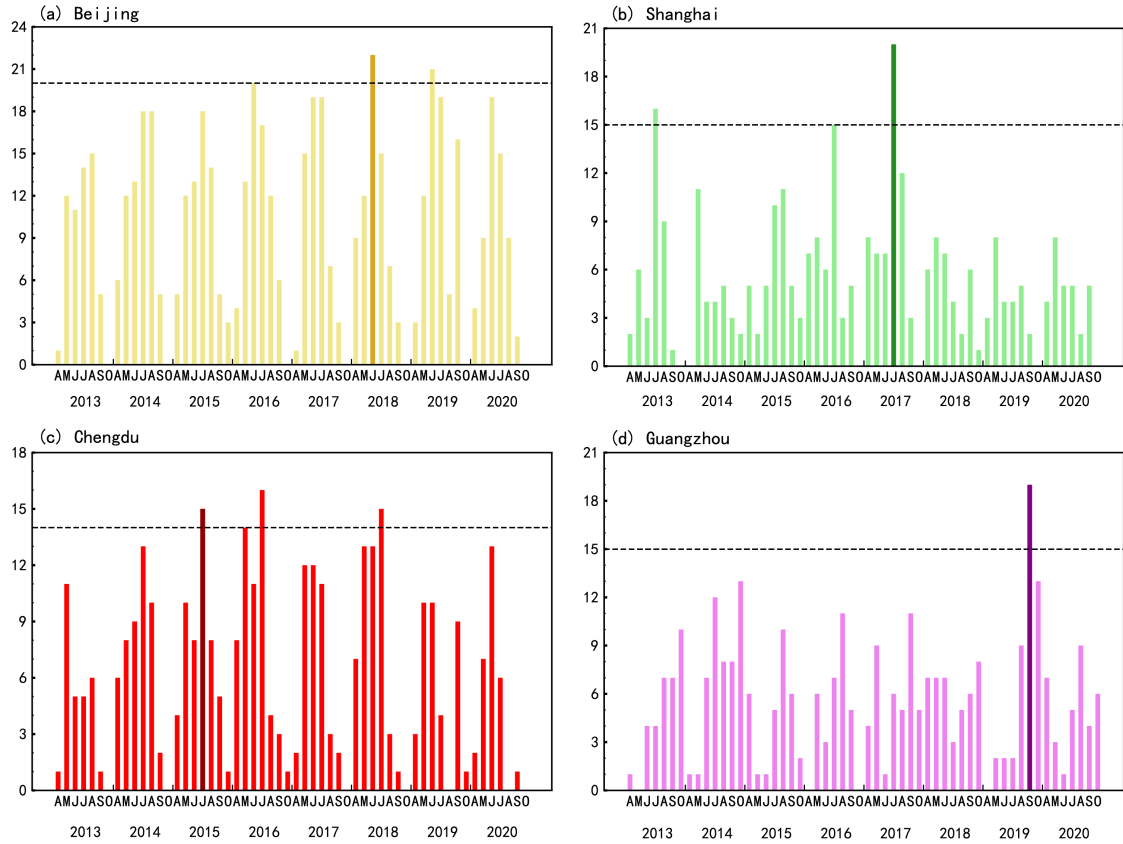
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592 **Table 2.** Horizontal mass transport (Tg) of O₃ from the surface to 500 hPa over NCP (115°–
593 120°E, 38°–44°N), YRD (120°–125°E, 28°–32°N), SCB (102.5°–105°E, 30°–32°N) and PRD
594 (110°–115°E, 22°–26°N) areas in the severe polluted months (June 2018, July 2017, July 2015
595 and September 2019, respectively) and averaged over the same months of a year during 1981–
596 2019, as well as their differences. Positive values indicate incoming fluxes and negative values
597 indicate outgoing fluxes.
598

| | Polluted month | Average | Anomalies |
|-------|----------------|---------|-----------|
| | NCP | | |
| North | 4.43 | 2.62 | 1.80 |
| South | -2.22 | -1.42 | -0.81 |
| East | -12.30 | -11.31 | -0.99 |
| West | 11.83 | 11.20 | 0.62 |
| | YPD | | |
| North | -4.13 | -3.88 | -0.25 |
| South | 3.58 | 3.20 | 0.37 |
| East | -2.05 | -3.90 | 1.85 |
| West | 2.03 | 4.04 | -2.01 |
| | SCB | | |
| North | 4.15 | 0.13 | 4.02 |
| South | -2.30 | 0.48 | -2.78 |
| East | -1.10 | -1.15 | 0.05 |
| West | 1.73 | 1.84 | -0.11 |
| | PRD | | |
| North | 2.70 | 0.72 | 1.97 |
| South | -2.87 | -0.90 | -1.96 |
| East | 2.24 | 1.15 | 1.09 |
| West | -2.32 | -1.55 | -0.76 |

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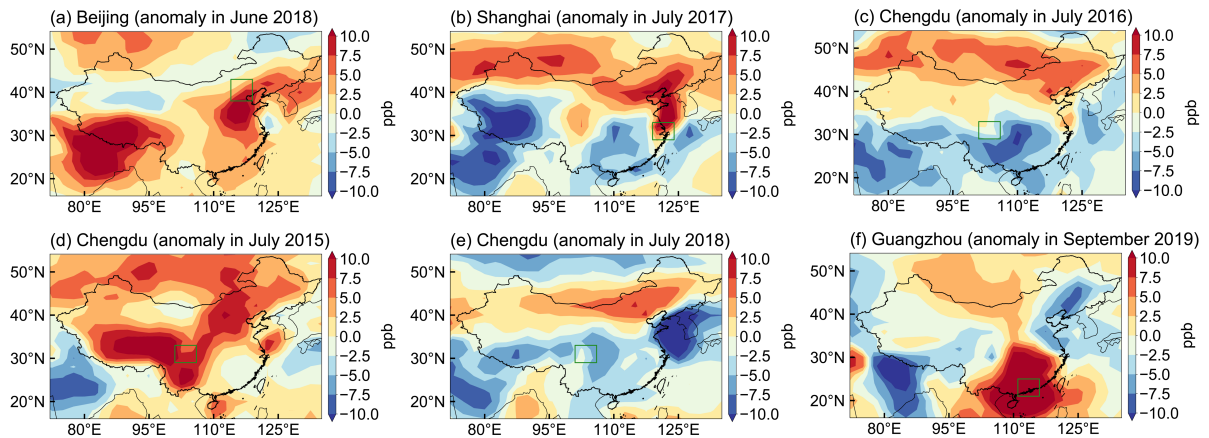
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Figure 1. Time series of frequencies of severe O₃ pollution days (defined by daily maximum of 8-h average ozone (MDA8-O₃) concentration greater than 160 μg m⁻³) in Beijing, Shanghai, Chengdu and Guangzhou (a–d) from April to ~~September~~ September–October during 2013–2020. The dark-colored bars represent the most severe month (second most for Chengdu) that has the highest frequency of O₃ pollution days for the individual cities.



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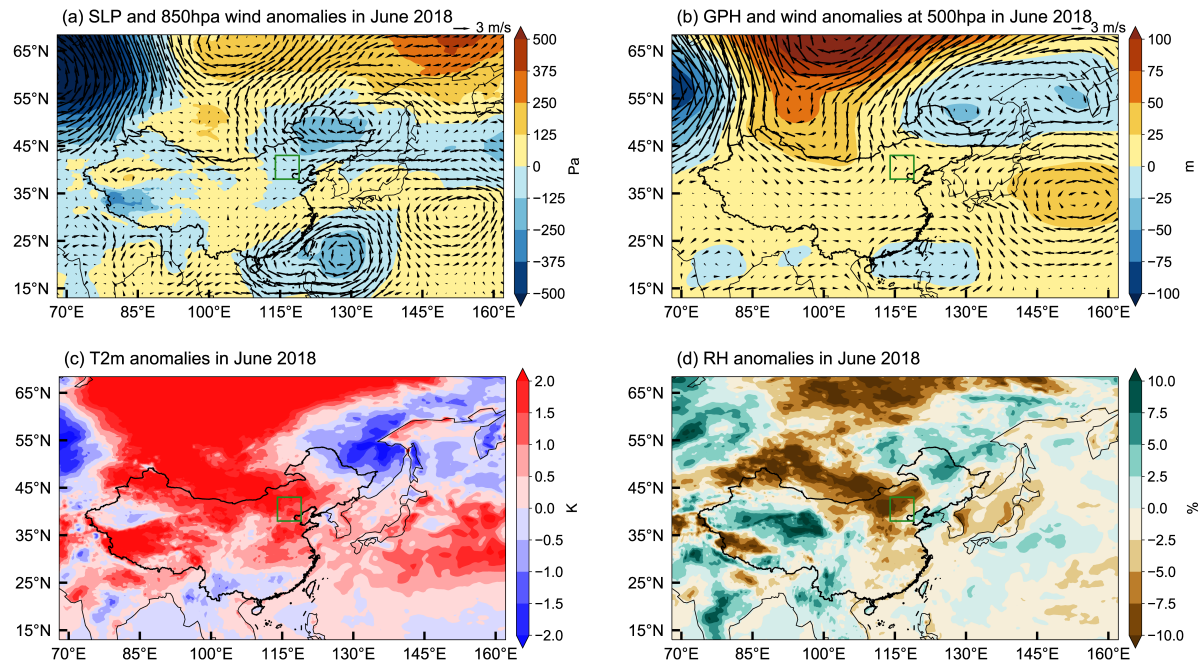
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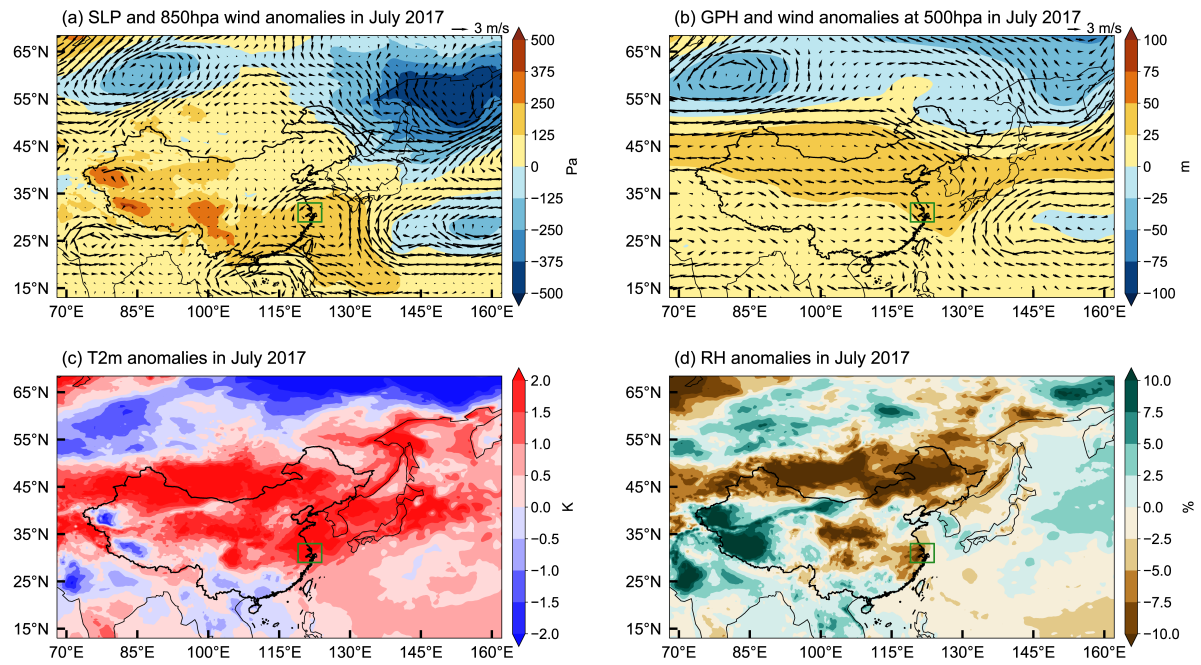
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Figure 2. Spatial distribution of monthly O₃ concentration anomalies (part per billion, ppb) in June 2018 (a), July 2017 (b), July 2016 (c), July 2015 (d), July 2018 (e) and September 2019 (f) relative to 40-year (1980–2019) monthly average for June (a), July (b, c, d, e) and September (f), simulated in the GEOS-Chem model. The green boxes mark NCP (a), YRD (b), SCB (c, d, e) and PRD (f). Anomalies are relative to the corresponding monthly averages over 1980–2019.



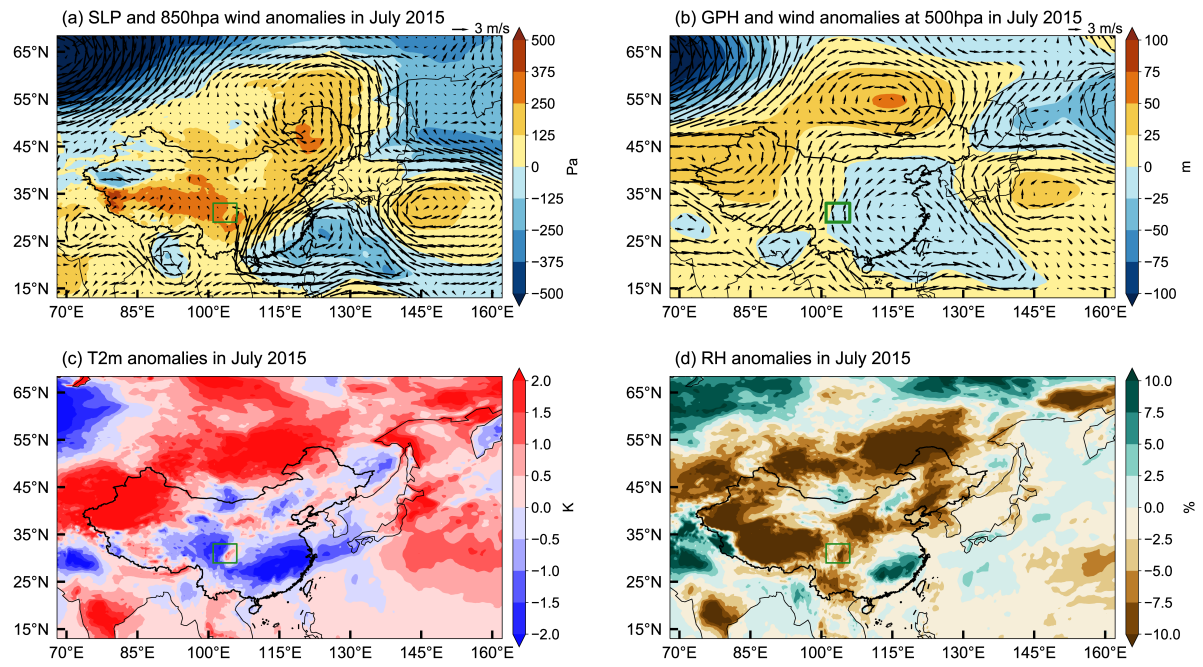
616

617 **Figure 3.** Anomalies in sea level pressure (SLP, Pa, shaded) and ~~1000~~1000-850 hPa winds (m s^{-1} ,
 618 vector) (a), geopotential height (GPH, m, shaded) and winds at 500 hPa (m s^{-1} , vector) (b), 2-
 619 meter air temperature (T2m, K) (c) and surface relative humidity (RH, %) (d) in June 2018
 620 relative to the 40-year (1980–2019) monthly average for June. The green boxes mark NCP.
 621



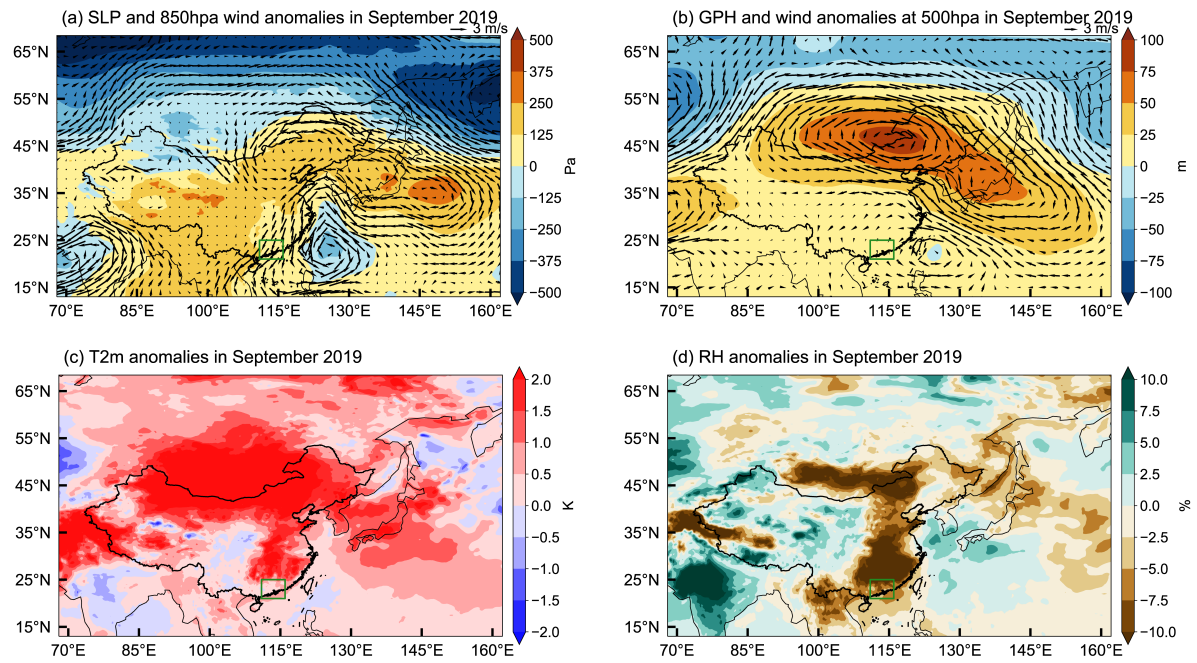
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Figure 4. Same as Figure 3 but for the monthly anomalies in July 2017. The green boxes mark YRD.



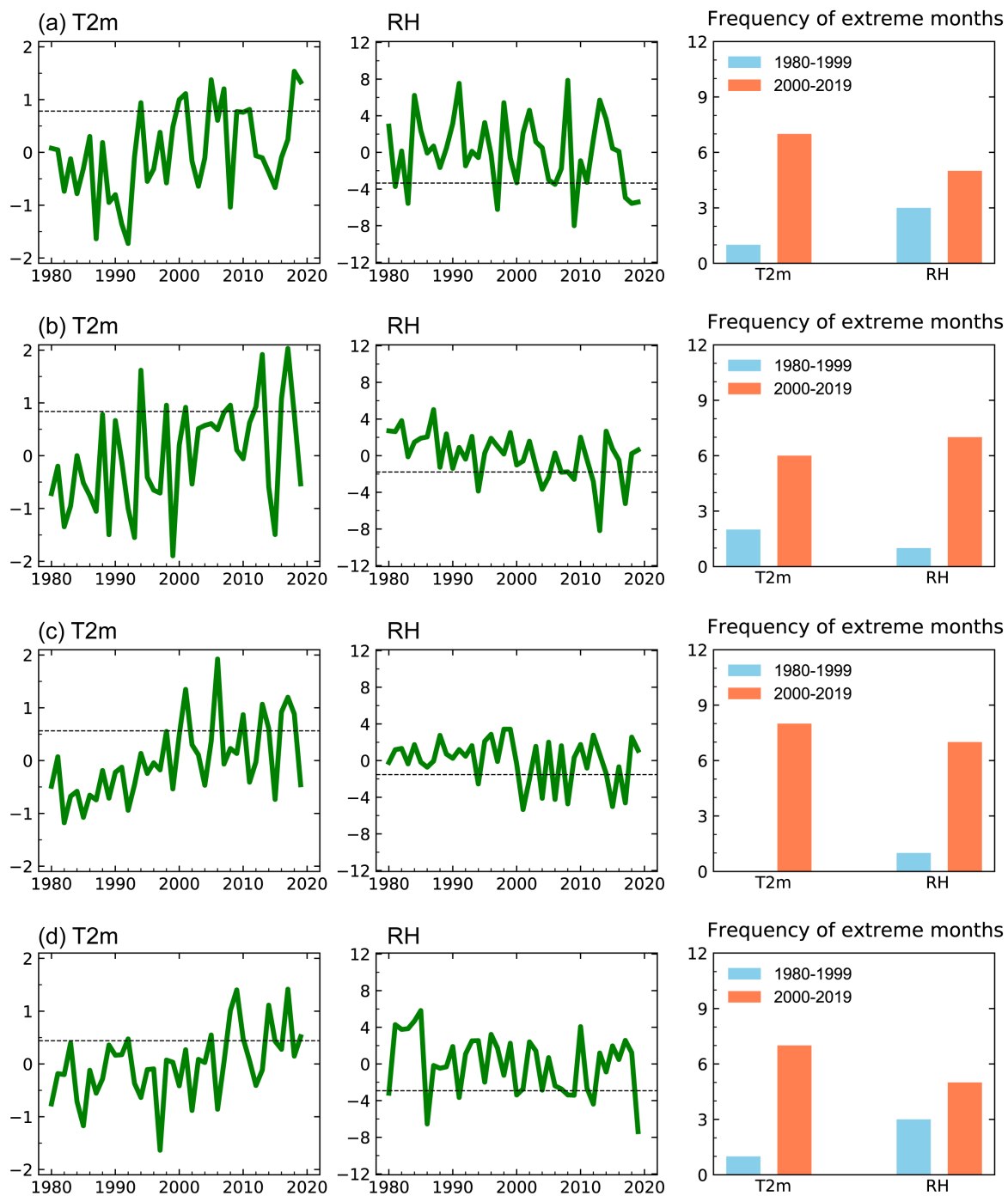
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Figure 5. Same as Figure 3 but for the monthly anomalies in July 2015. The green boxes mark SCB.



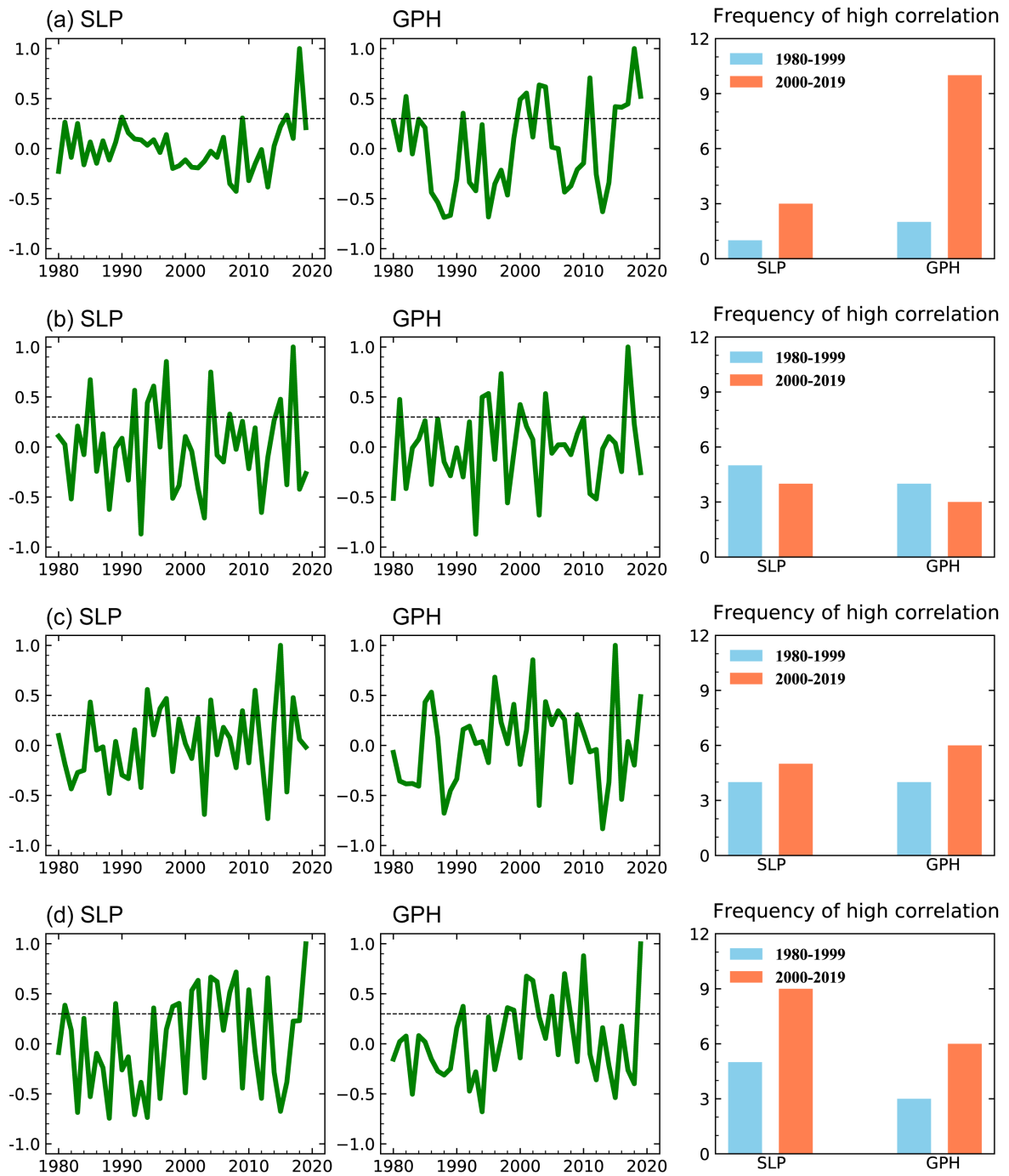
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631 **Figure 6.** Same as Figure 3 but for the monthly anomalies in September 2019. The green boxes
 632 mark PRD.



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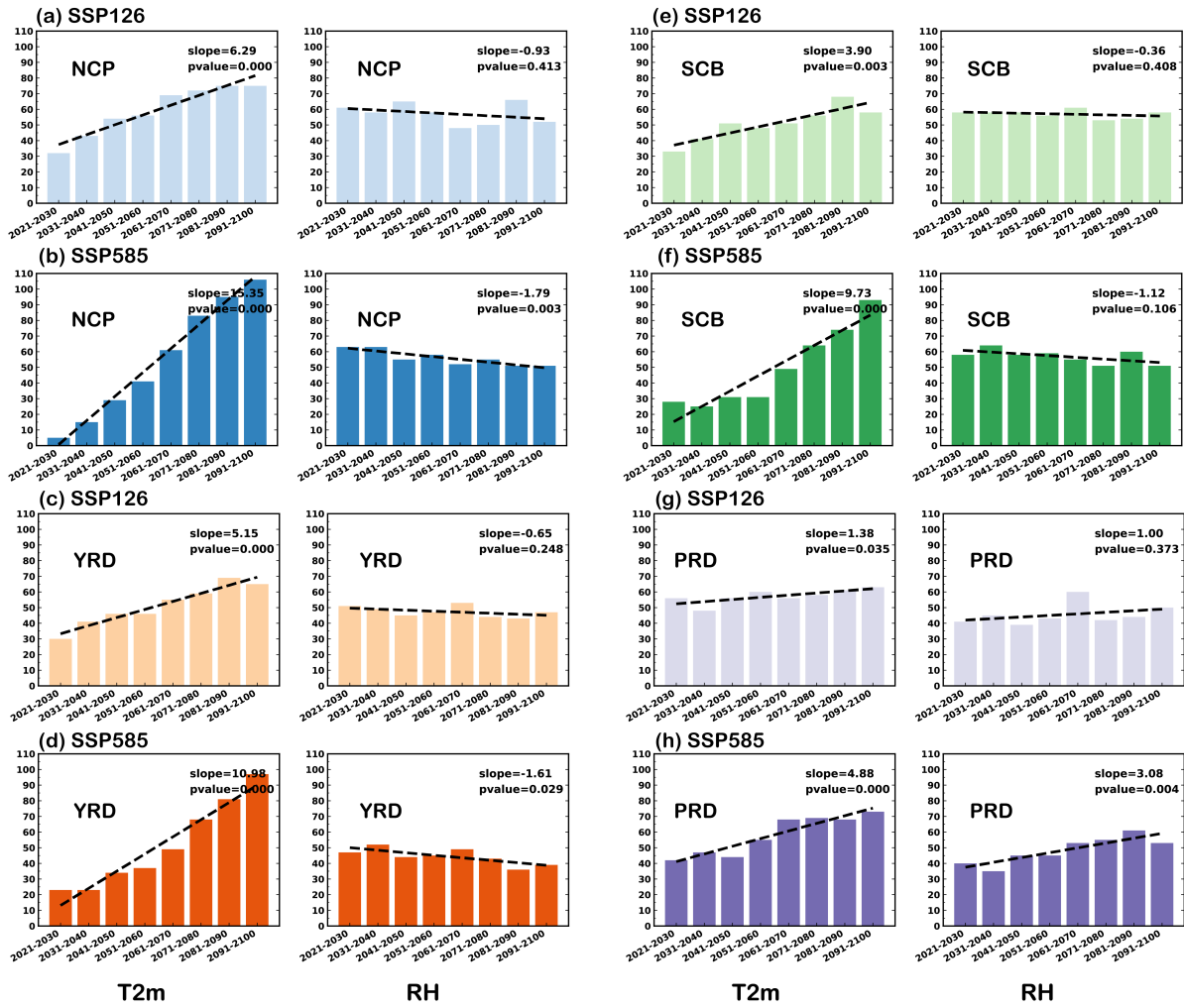
634 **Figure 7.** Time series of anomalies of T2m (K, left) and surface RH at 1000 hPa (% , middle)
 635 over (a) NCP (115°–120°E, 38°–44°N), (b) YRD (120°–125°E, 28°–32°N), (c) SCB (102.5°–
 636 105°E, 30°–32°N) and (d) PRD (110°–115°E, 22°–26°N) in the most polluted months during
 637 1980–2019. The dotted lines mark the 80th percentile of the distributions for T2m and 20th
 638 percentile for RH. The bar charts (right) represent the frequency of T2m above the 80th
 639 percentile and RH anomalies below the 20th percentile during 1980–1999 (blue) and 2000–
 640 2019 (orange).



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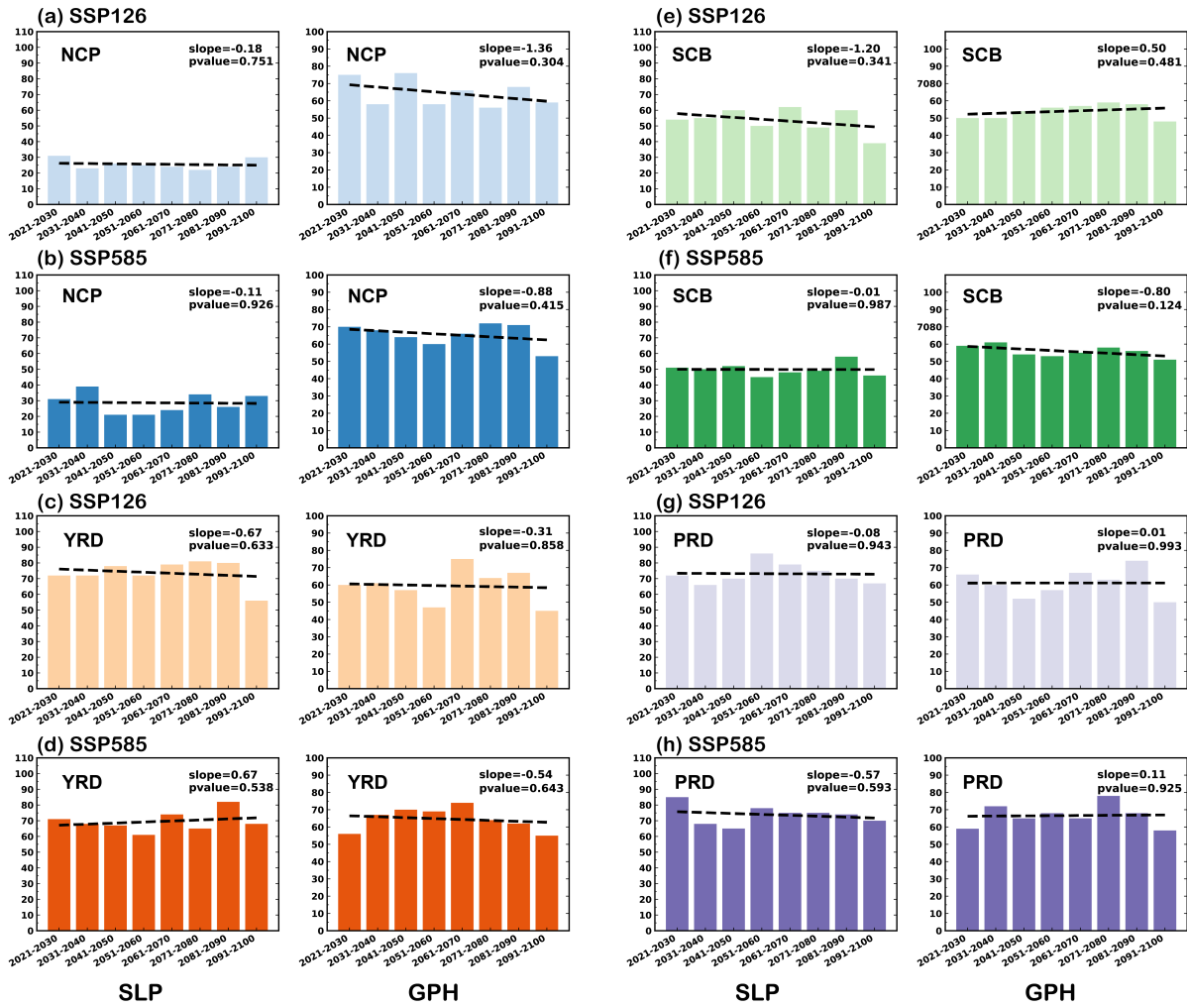
642 **Figure 8.** Time series of spatial correlation in SLP (left) and 500 hPa GPH (middle) anomalies
 643 over East Asia and Western Pacific (EAWP, 90°–160°E, 20°–60°N) in June 2018 (a), July 2017
 644 (b), July 2015 (c) and September 2019 (d) and those in the same targeted month of each year
 645 during 1980–2019. The dotted lines mark the correlation coefficient of +0.3, which is used as
 646 a threshold to define “moderate to high correlation”. The bar chart (right) represents the
 647 frequency of SLP and 500 hPa GPH anomalies in the same months during 1980–1999 (blue)
 648 and 2000–2019 (orange) that have moderate to high correlation (>0.3) with those in June 2018,
 649 July 2017, July 2015 and September 2019.

650



651

652 **Figure 9.** Frequencies of extreme months with T2m or RH anomalies exceeding the 80th
 653 percentile or below the 20th percentile of the distributions over NCP (115°–120°E, 38°–44°N)
 654 (a, b), YRD (120°–125°E, 28°–32°N) (c, d), SCB (102.5°–105°E, 30°–32°N) (e, f) and PRD
 655 (110°–115°E, 22°–26°N) (g, h) in each 10-year interval during 2021–2100 under two SSPs
 656 future scenarios of 13 CMIP6 models. The two SSPs are SSP1-2.6 and SSP5-8.5. The slope
 657 and P values of the linear regression during 2021–2100 are shown in the upper right of each
 658 panel.



659

660 **Figure 10.** Frequencies of extreme months with SLP and 500 hPa GPH that have moderate to
 661 high correlation (>0.3) to those in June 2018 (a, b), July 2017 (c, d), July 2015 (e, f) and
 662 September 2019 (g, h) in each 10-year interval during 2021–2100 under two SSPs future
 663 scenarios of 13 CMIP6 models. The two SSPs are SSP1-2.6 and SSP5-8.5. The slope and P
 664 values of the linear regression during 2021–2100 are shown in the upper right of each panel.
 665 The linear trends of SLP and GPH in each model grid were removed before the correlation
 666 coefficient is calculated.