



1	Meteorological characteristics of severe ozone pollution events in
2	China and their future predictions
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### 23 Abstract

24	Ozone (O <sub>3</sub> ) 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, meteorological characteristics conducive to severe O <sub>3</sub> pollution
27	in various regions of China are investigated, and their historical changes and future trends are
28	analyzed. During the most severe O <sub>3</sub> pollution months over the North China Plain (NCP) and
29	Yangtze River Delta (YRD), the chemical production of O <sub>3</sub> is enhanced under the hot and dry
30	conditions, while the regional transport is the main reason causing the severe O <sub>3</sub> pollution over
31	Sichuan Basin (SCB) and Pearl River Delta (PRD) during the severe polluted months. Over
32	the last four decades, the frequencies of high temperature and low relative humidity conditions
33	increased in 2000-2019 relative to 1980-1999, indicating that O <sub>3</sub> pollution in both NCP and
34	YRD became more frequent under the historical climate change. In SCB and PRD, the
35	occurrence of atmospheric circulation patterns similar to those during the polluted months
36	increased, together with the more frequent hot and dry conditions, contributing to the increases
37	in severe O3 pollution in SCB and PRD during 1980–2019. In the future (by 2100), the
38	frequencies of months with anomalous high temperature show stronger increasing trends in the
39	high forcing scenario (SSP5-8.5) compared to the sustainable scenario (SSP1-2.6) in China. It
40	suggests that high anthropogenic forcing will not only lead to slow economic growth and
41	climate warming, but also likely result in environmental pollution issues.





# 42 1. Introduction

43 Tropospheric ozone  $(O_3)$ , one major air pollutant, is formed in photochemical reactions of nitrogen oxides (NO<sub>x</sub>) and volatile organic compounds (VOCs) when exposed to sunlight 44 (Finlayson-Pitts and Pitts, 1997; Silman, 1999). Enhanced O<sub>3</sub> pollution harms ecosystems and 45 human health (Fleming et al., 2018; Maji et al., 2019) by reducing crop yields (Ainsworth et 46 al., 2012; Mills et al., 2018) and aggravating cardiopulmonary disease (Ebi and McGregor, 47 2008; Liu et al. 2018). In recent years, near-surface ozone concentrations in many regions of 48 49 China have been increasing considerably (Verstraeten et al., 2015; Cheng et al., 2019; Zhang 50 et al., 2020, Li et al., 2019; Lu et al., 2018; Silver et al., 2018; Yin et al., 2019, Lu et al., 2020). 51 Lu et al. (2020) revealed that the daily maximum of 8-h average O<sub>3</sub> concentration (MDA8-O<sub>3</sub>) in China increased by 2.4 ppb per year (5.0% relative to the average) during April–September 52 over 2013-2019. 53

54 In addition to emissions, O<sub>3</sub> concentrations are influenced by meteorological factors such as temperature, relative humidity, solar radiation, and winds (Mott et al., 2005; Fu and Tian, 55 2019; Gong and Liao, 2019; Li et al., 2019, 2020; Le et al., 2020; Zhao et al., 2020). Typically, 56 57 strong solar radiation, high surface air temperatures, and low relative humidity are conducive 58 to photochemical production of O<sub>3</sub>, causing a raise of O<sub>3</sub> concentration (Peterson and Flowers, 1977; Xu, et al., 2011; Coates et al., 2016; Li et al., 2020; Dang et al., 2021). Wind speed is 59 60 negatively correlated with surface  $O_3$  because low wind speed facilitates the accumulation of 61 O<sub>3</sub> upon production (Zhang et al., 2015; Wang et al., 2017; Liu and Wang, 2020). Han et al. (2020) explored the impacts of various meteorological factors on the daily variation of summer 62 63 surface  $O_3$  in eastern China based on a multiple linear regression method and suggested that





relative humidity is the primary factor affecting O<sub>3</sub> concentration in central and south parts of eastern China, while temperature is the most important factor governing O<sub>3</sub> concentration in north of eastern China. Gong and Liao (2019) reported that the meteorological characteristics of O<sub>3</sub> pollution events in North China during 2014–2017 were the high daily maximum temperature, low relative humidity, abnormal southerly winds and high pressure at 500 hPa. These findings emphasize that meteorological factors play a crucial role in regulating O<sub>3</sub> pollution in China.

71 Atmospheric circulation patterns affect O<sub>3</sub> concentrations over China through changing meteorological factors (Yang et al, 2014; Zhao and Wang, 2017; Shu et al., 2019; Dong et al., 72 2020; Zhou et al., 2022). Zhao and Wang (2017) examined the influence of the Western Pacific 73 Subtropical high (WPSH) on O<sub>3</sub> over eastern China based on observations and reanalysis data 74 from 2014 to 2016. They found that stronger WPSH enhanced the moisture transport to 75 76 southern China, which was detrimental to the photochemical reaction of O<sub>3</sub>, leading to a decrease in surface O<sub>3</sub> concentration in southern China, whereas O<sub>3</sub> concentrations in northern 77 China increased under the stronger WPSH related to the dry and hot conditions favoring O<sub>3</sub> 78 79 production. On the basis of observational O<sub>3</sub> data and ERA5 reanalysis data during 2014–2018, 80 Dong et al. (2019) analyzed the impact of synoptic patterns on summertime O<sub>3</sub> pollution in the North China Plain and revealed that the most severe O<sub>3</sub> pollution weather pattern is associated 81 82 with anomalous southwesterly winds, which carry dry, warm air from inland southern China 83 to the North China Plain and favor the chemical production of O<sub>3</sub>. Zhou et al. (2022) explored 84 the impacts of Asian summer monsoon on the interannual variation of O<sub>3</sub> concentrations based 85 on surface measurements and GEOS-Chem model simulations. They showed that the East





- Asian summer monsoon strength was positively correlated with O<sub>3</sub> concentration in southcentral China and South Asian summer monsoon has complex effects on O<sub>3</sub> pollution in China,
- 88 mainly through changing transboundary transport related to large-scale circulations.
- 89 As mentioned above, previous studies have examined the meteorological characteristics 90 of O<sub>3</sub> pollution in limited regions in China. In this study, the meteorological characteristics 91 conducive to severe O<sub>3</sub> pollution in several polluted areas of China, including the North China 92 Plain (NCP), Yangtze River Delta (YRD), Sichuan Basin (SCB), and Pearl River Delta (PRD), 93 are investigated based on the observed surface O<sub>3</sub> concentrations, reanalysis data, and GEOS-94 Chem model simulations. Besides, the contributions from various chemical and physical 95 processes inducing regional O<sub>3</sub> pollution are quantified using an integrated process rate (IPR) 96 analysis method. Moreover, variations of future meteorological patterns leading to severe  $O_3$ 97 pollution in China are presented under the sustainable and high forcing scenarios according to 98 the multi-model data from the Coupled Model Intercomparison Project Phase 6 (CMIP6).

# 99 **2. Methods**

#### 100 2.1 Surface ozone observations and meteorological reanalysis

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





108	The meteorological fields are taken from the European Centre for Medium-Range
109	Weather Forecasts (ECMWF) ERA5 monthly reanalysis dataset during 1980-2020, with a
110	horizontal resolution of $0.25^{\circ} \times 0.25^{\circ}$ . To explore the meteorological characteristics that are
111	conducive to O3 pollution, sea level pressure (SLP), geopotential height (GPH) at 500 hPa,
112	wind fields at 850 hPa and 500 hPa, temperature at 2m (T2m) and surface relative humidity
113	(RH) are adopted, which can have significant impacts on O <sub>3</sub> variations in China (Jiang et al.,
114	2020; Dong et al., 2020; Le et al., 2020).

#### 115 2.2 GEOS-Chem model simulations

116 O<sub>3</sub> concentrations and the related chemical and physical processes causing O<sub>3</sub> variations over 1981-2020 are simulated in the global atmospheric chemistry model GEOS-Chem 117 (version V12.9.3), driven by the Modern-Era Retrospective analysis for Research and 118 Application, Version 2 (MERRA-2). Simulations are performed on 47 vertical layers from 119 120 surface to 0.01 hPa, and a horizontal grid of  $2^{\circ}$  latitude  $\times 2.5^{\circ}$  longitude. GEOS-Chem model incorporates a fully coupled  $O_3$ -NO<sub>x</sub>-hydrocarbon-aerosol chemical mechanism (Pye et al., 121 122 2009; Mao et al., 2013; Sherwen et al., 2016). Boundary-layer mixing uses a non-local scheme 123 (Lin and McElroy, 2010), and stratospheric O<sub>3</sub> chemistry employs the linearized O<sub>3</sub> parameterization (LINOZ) (McLinden et al., 2000). 124

Global anthropogenic aerosol and precursor gas emissions driving the simulations are from the Community Emissions Data System (CEDS, Hoesly et al., 2018) and biomass burning emissions are from the Global Fire Emissions Database, Edition 4 (GFED4, Van der Werf et al., 2017). VOCs emissions from biogenic sources are provided offline by the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN V2.1, Guenther et al.,





130	2012). Lightning and soil emissions are specified in the model (Hudman et al., 2012; Ott et al.,
131	2010). Anthropogenic emissions in China are updated with the Multi-resolution Emission
132	Inventory (MEIC), a localized emission dataset for China. Anthropogenic, biomass burning,
133	biological and other natural emissions are kept at 2017 level during the simulations, so as to
134	eliminate the influence of emission changes on the interannual variation and trends of $O_3$ .
135	Simulated O <sub>3</sub> distributions with the same configuration in GEOS-Chem have been extensively
136	evaluated in many studies, and the model has been reported to capture O3 concentrations well
137	in China (e.g., Li et al., 2019; Lu et al., 2019; Ni et al., 2018).

## 138 2.3 CMIP6 multi-model simulations

The multi-model simulations from historical and the Scenario Model Intercomparison 139 Project (ScenarioMIP) in CMIP6 are used to analyze the historical variations and future trends 140 of meteorological conditions conducive to severe O<sub>3</sub> pollution. Two different future scenarios 141 142 of the Shared Socioeconomic Path (SSPs) are applied, including the sustainable scenario (SSP1-2.6) and the high forcing scenario (SSP5-8.5). Totally simulations from 13 models 143 (ACCESS-CM2, ACCESS-ESM1-5, CAS-ESM2-0, CMCC-CM2-SR5, CMCC-ESM2, 144 145 FGOALS-f3-L, FGOALS-g3, GFDL-ESM4, INM-CM4-8, INM-CM5-0, IPSL-CM6A-LR, MPI-ESM1-2-HR, MPI-ESM1-2-LR) are analyzed in this study. 146

## 147 **3. Results**

#### 148 **3.1 Meteorological characteristics conductive to regional ozone pollution**

To investigate the relationship between meteorological conditions and regional O<sub>3</sub> pollution in China, the frequencies of O<sub>3</sub> pollution days from April to September during 2013– 2020 are calculated for Beijing, Shanghai, Chengdu and Guangzhou, representing the typical





152	four polluted regions in China (i.e., NCP, YRD, SCB and PRD) (Figure 1). Observational data
153	show the highest frequencies of $O_3$ pollution days in June 2018, July 2017 and September 2019
154	in Beijing, Shanghai and Guangzhou, with pollution days up to 22, 20 and 19 days per month,
155	respectively. Variations in O <sub>3</sub> concentration in the real world are driven by changes in both
156	meteorological factors and emissions. With fixed emissions, the positive anomalies of near-
157	surface O3 concentrations over NCP, YRD and PRD during their most polluted months can also
158	be reproduced by the GEOS-Chem model (Figure 2), suggesting that the O <sub>3</sub> pollutions during
159	the most polluted months over NCP, YRD and PRD are likely attributable to the anomalies of
160	meteorological conditions. In the top three O <sub>3</sub> polluted months in Chengdu, only in July 2015
161	the higher concentrations than the long-term averages can be captured by the ssimulations with
162	fixed emissions. Therefore, in this study, we focus on the meteorological characteristics in June
163	2018, July 2017, July 2015 and September 2019, that were conducive to the severe $O_3$ pollution
164	over NCP, YRD, SCB and PRD, respectively.

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When O<sub>3</sub> pollution was the most severe over NCP in June 2018, an anomalous high 165 pressure occurred at 500 hPa over NCP (Fig. 3b), relative to the 40-year climatological 166 167 averages from 1980 to 2019, leading to positive T2m anomalies near the surface (Fig. 3c). Anomalous lows located over northeastern China and northwestern Pacific (Fig. 3a) and the 168 associated anomalous northerly winds prevent the moisture moving from the ocean to NCP, 169 causing negative RH anomalies over NCP (Fig. 3d). The meteorological conditions with the 170 171 high T2m and low RH are favorable for the photochemical production of O3. When the most severe O3 pollution occurred in July 2017, YRD was dominated by anomalous high pressure 172 in the lower and middle troposphere (Figs. 4a and 4b). Under the control of high pressure, the 173

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174	meteorological conditions (e.g., high T2m and low RH) enhance the photochemical production
175	of $O_3$ (Figs. 4c and 4d). In the $O_3$ pollution event of SCB in July 2015, the negative T2m
176	anomaly is not conducive to the O <sub>3</sub> production (Fig. 5c), although the RH was low (Fig. 5d).
177	Meanwhile, the anomalous low over eastern China and northwestern Pacific in the middle
178	troposphere favors regional O3 transport from the polluted source region over eastern China to
179	SCB (Fig. 5b) and the anomalous high over central-western China is conducive to the vertical
180	transport of upper tropospheric $O_3$ down to the lower troposphere (Fig. 5a). For the PRD in
181	September 2019, the anomalous high covering almost the entire China along with the
182	anomalous low over East China Sea generates northerly wind anomalies in the lower
183	troposphere over eastern China, which tend to transport polluted air from northern China and
184	weaken the inflow of oceanic clean air (Fig. 6). The temperature increase is much more
185	significant in the upwind regions as compared to PRD, suggesting that the strong regional
186	transport could be the primary reason causing this severe O <sub>3</sub> pollution event of PRD.

## 187 **3.2** Physical and chemical mechanisms leading to regional ozone pollution

To further explore the mechanisms of meteorological changes leading to the severe O<sub>3</sub> pollution over the four typical polluted regions in China, contributions of individual chemical and physical processes to O<sub>3</sub> variations are quantified based on the IPR analysis from GEOS-Chem simulations and summarized in Table 1.

192 Consistent with the meteorological anomalies analyzed above, high temperature and low 193 RH meteorological conditions in NCP are conducive to the photochemical production of O<sub>3</sub>. 194 During the polluted month over NCP, the chemical production of tropospheric O<sub>3</sub> is higher than 195 the long-term average by 2.36 Gg day<sup>-1</sup>, while the horizontal transport also contributes to the





196	increase in $O_3$ mass by 1.58 Gg day <sup>-1</sup> (Table 1). Due to the enhanced northwesterly winds, the
197	import of $O_3$ mass from the north and east of NCP was increased by 1.80 and 0.62 Tg,
198	respectively (Table 2). In YRD, the chemical production (2.38 Gg day <sup>-1</sup> ) is also the dominant
199	process that drives the O <sub>3</sub> concentration increase during the severe polluted month, associated
200	with the warm and dry conditions. Therefore, the anomalous chemical production is the major
201	process that induced O <sub>3</sub> pollution in NCP and YRD during the severe polluted months.
202	Different from NCP and YRD, horizontal transport is the main process that caused O <sub>3</sub>
203	pollution in SCB and PRD during the severe polluted months. It contributes to the rate of
204	increase in $O_3$ mass by 5.10 and 6.67 Gg day <sup>-1</sup> , respectively, over SCB and PRD, while other
205	processes tend to decrease the O <sub>3</sub> mass (Table 1). Due to the anomalous northerly winds over
206	SCB, more $O_3$ is transported into SCB from north (by 4.02 Tg), and the anomalous
207	northeasterly winds enhance the $O_3$ transport from the north and east of PRD by 1.97 and 1.09
208	Tg, respectively, leading to the increase in O <sub>3</sub> concentrations over SCB and PRD during the
209	severe polluted months relative to the climatological averages (Table 2).

## 210 **3.3 Historical and future changes in the meteorological conditions**

O<sub>3</sub> pollution has deteriorated in China during recent decades, which could be related to the changes in meteorological conditions. Time series of T2m and RH anomalies in the polluted months during the 1980–2019 and frequencies of high T2m and low RH months during 1980– 1999 and 2000–2019 over the four polluted regions in China based on ERA5 reanalysis data are shown in Figure 7. Due to climate change, both the high temperature and low RH conditions in NCP, YRD, SCB and PRD all increased during the past four decades (2000-2019 versus 1980-1999). Based on the analysis showing that chemical production is the dominant process





218	of severe O <sub>3</sub> pollution in NCP and YRD, the increases in the frequency of high temperature
219	and low RH indicate that severe O3 pollution in both NCP and YRD has become more frequent
220	under the historical climate change. In SCB and PRD, the severe O <sub>3</sub> pollution is more related
221	to changes in regional transport. The SLP and 500 hPa GPH over East Asia and Western Pacific
222	similar to those during the severe polluted months in both SCB and PRD have increased (2000-
223	2019 versus 1980-1999) (Figure 8), together with the more frequent hot and dry conditions
224	(Figure 7), leading to the increases in severe O <sub>3</sub> pollution in SCB and PRD during 1980–2019.
225	Many studies have reported that future climate change will have significant influences
226	on O <sub>3</sub> pollution in China through changing meteorological factors (e.g., Li et al., 2023; Wang
227	et al., 2022). Here, the frequencies of extreme months with high T2m and low RH and the
228	frequencies of extreme months with SLP and 500 hPa GPH that have moderate to high
229	correlation to those in the polluted months in the four regions of China, under the sustainable
230	(SSP1-2.6) and high forcing (SSP5-8.5) scenarios during 2021–2100 from CMIP6 multi-model
231	results, are presented in Figures 9 and 10, respectively. The frequencies of months with
232	anomalous high temperature show obvious upward trends in both SSP1-2.6 and SSP5-8.5
233	scenarios over the four regions, and the increasing trends in SSP5-8.5 are much more
234	significant than in SSP1-2.6. Frequencies of low RH months show downward trends in NCP,
235	YRD and SCB, especially under SSP5-8.5, while there is an upward trend in PRD. Note that
236	the trends in frequencies of low RH months are much less significant than in high temperature
237	months. The frequencies of extreme months with SLP and 500 hPa GPH that are similar to
238	those in the severe O <sub>3</sub> pollution months in the four regions do not show significant trends in
239	the SSPs. Hence, the future climate change may aggregate O <sub>3</sub> pollution in China by enhancing





the chemical production related to temperature increases. The O<sub>3</sub> pollution exacerbation is projected to be less significant in the sustainable scenario due to the moderate temperature increase than in the high forcing scenario, suggesting that the sustainable scenario is the optimal path to retaining clean air in China. High anthropogenic radiative forcing will not only lead to slow economic growth and climate warming, but also result in the environmental pollution.

### **4.** Conclusions

247 O<sub>3</sub> pollution harms ecosystems and human health. In recent years, near-surface O<sub>3</sub> concentrations in many regions of China have been increasing considerably. Base on 248 observational O<sub>3</sub> data, ERA5 reanalysis data and GEOS-Chem model simulations, 249 meteorological characteristics conducive to severe O<sub>3</sub> pollution in different regions of China 250 are investigated in this study. Contributions from various chemical and physical processes 251 252 inducing O<sub>3</sub> pollution are quantified using an integrated process rate (IPR) analysis method. Furthermore, historical changes and future trends of meteorological conditions leading to 253 severe O<sub>3</sub> pollution in China are explored based on the meteorological reanalysis and CMIP6 254 255 multi-model future predictions, which is of great implication for the mitigation and prevention 256 of O<sub>3</sub> pollution over China.

In this study, June 2018, July 2017, July 2015 and September 2019 are identified as the most severe  $O_3$  pollution months influenced by meteorological factors over NCP, YRD, SCB and PRD, respectively. Severe  $O_3$  pollution in June 2018 over NCP and in July 2017 over YRD is mainly due to enhanced chemical production related to hot and dry conditions. The chemical production of  $O_3$  in the severe polluted months over NCP and YRD are 2.36 Gg day<sup>-1</sup> and 2.38





262	Gg day <sup>-1</sup> , respectively, higher than the climatological averages. Different from NCP and YRD,
263	regional transport is the main process leading to the high O <sub>3</sub> concentration in SCB and PRD
264	during the respective severely polluted months, which contributes to the rate of increase in $O_3$
265	mass by 5.10 and 6.67 Gg day <sup>-1</sup> , respectively, over SCB and PRD. During the severely polluted
266	months, related to large-scale circulation patterns, anomalous northerly winds transport more
267	O3 into SCB from north, and anomalous northeasterly winds enhance the O3 transport from the
268	north and east into PRD.

269 Over the last four decades (2000-2019 versus 1980-1999), the frequencies of high 270 temperature and low RH increased, indicating that O<sub>3</sub> pollution in both NCP and YRD has become more frequent under the historical climate change. In SCB and PRD, the occurrence 271 272 of atmospheric circulation patterns similar to those during the polluted months in both SCB 273 and PRD has increased, together with the more frequent hot and dry conditions, leading to the 274 increases in severe O<sub>3</sub> pollution in SCB and PRD during 1980–2019. In the future (by 2100), 275 the frequencies of months with anomalous high temperature show obvious upward trends in 276 both sustainable (SSP1-2.6) and high forcing (SSP5-8.5) scenarios over the four regions, and 277 the increasing trends in SSP5-8.5 are much more significant than in SSP1-2.6. This suggests that high anthropogenic radiative forcing will not only lead to slow economic growth and 278 279 climate warming, but also likely result in environmental pollution issues. The sustainable scenario is the optimal path to retaining clean air in China. 280





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516	Code and data availability. The GEOS-Chem model is available at
517	https://zenodo.org/record/3974569#.YTD81NMzagR (last access: June 2023). O3 observations
518	over China can be obtained at https://quotsoft.net/air (last access: June 2023). ERA5 reanalysis
519	data can be downloaded at https://www.ecmwf.int/en/forecasts/datasets/reanalysis-
520	datasets/era5 (last access: June 2023). The multi-model simulations of the Coupled Model
521	Intercomparison Project Phase 6 (CMIP6) are from https://esgf-node.llnl.gov/search/cmip6/
522	(last access: June 2023).
523	Author contribution. YY designed the research; YY and YZ performed simulations and
524	analyzed the data. All authors including HW, LH, PW, and HL discussed the results and wrote
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537	Table 1. Anomalies in net rate of changes in tropospheric O <sub>3</sub> mass (Gg day <sup>-1</sup> ) over NCP (115°-
538	120°E, 38°–44°N), YRD (120°–125°E, 28°–32°N), SCB (102.5°–105°E, 30°–32°N) and PRD

 $(110^{\circ}-115^{\circ}E, 22^{\circ}-26^{\circ}N)$  due to physical and chemical processes in the most polluted months

540 (June 2018, July 2017, July 2015 and September 2019, respectively) relative to the same

541 months averaged during 1981–2019.

542

	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

543





Table 2. Horizontal mass transport (Tg) of O3 from the surface to 500 hPa over NCP (115°-545

120°E, 38°-44°N), YRD (120°-125°E, 28°-32°N), SCB (102.5°-105°E, 30°-32°N) and PRD 546

(110°-115°E, 22°-26°N) areas in the severe polluted months (June 2018, July 2017, July 2015 547

and September 2019, respectively) and averaged over the same months of a year during 1981-548

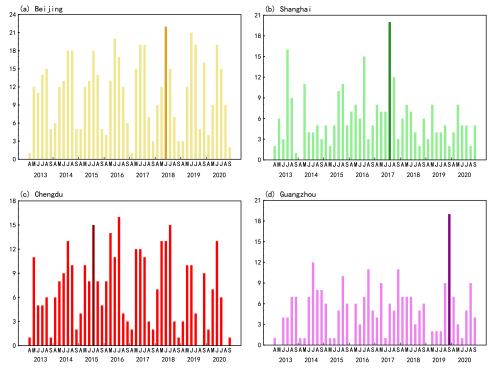
2019, as well as their differences. Positive values indicate incoming fluxes and negative values 549

550 indicate outgoing fluxes. 551

	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





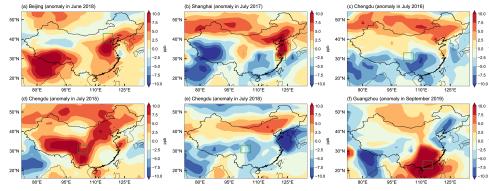


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**Figure 1.** Time series of frequencies of severe  $O_3$  pollution days (defined by daily maximum of 8-h average ozone (MDA8-O<sub>3</sub>) concentration greater than 160 µg m<sup>-3</sup>) in Beijing, Shanghai, Chengdu and Guangzhou (a–d) from April to September during 2013–2020. The dark-colored bars represent the most severe month (second most for Chengdu) that has the highest frequency of O<sub>3</sub> pollution days for the individual cities.







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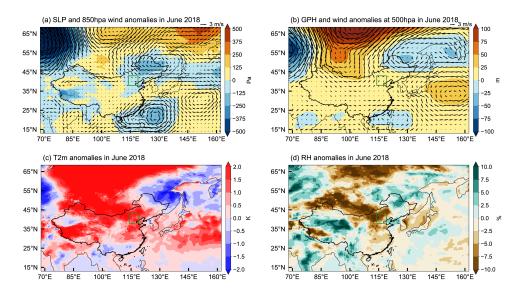
562 **Figure 2.** Spatial distribution of monthly  $O_3$  concentration anomalies (part per billion, ppb) in

June 2018 (a), July 2017 (b), July 2016 (c), July 2015 (d), July 2018 (e) and September 2019 (f), 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.





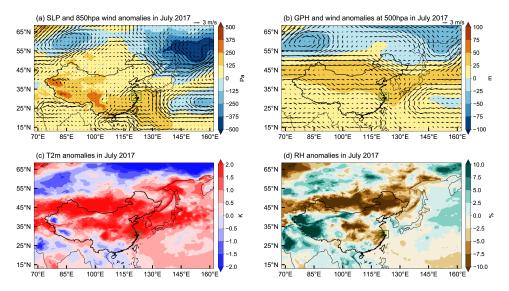


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Figure 3. Anomalies in sea level pressure (SLP, Pa, shaded) and 1000 hPa winds (m s<sup>-1</sup>, vector)
(a), geopotential height (GPH, m, shaded) and winds at 500 hPa (m s<sup>-1</sup>, vector) (b), 2-meter air
temperature (T2m, K) (c) and surface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





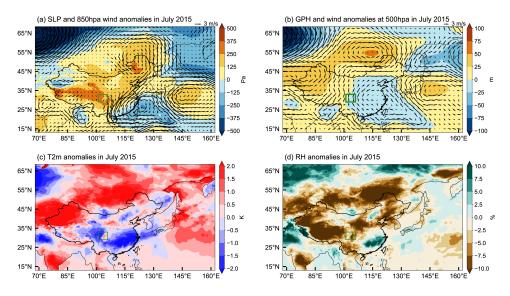


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Figure 4. Same as Figure 3 but for the monthly anomalies in July 2017. The green boxes markYRD.







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





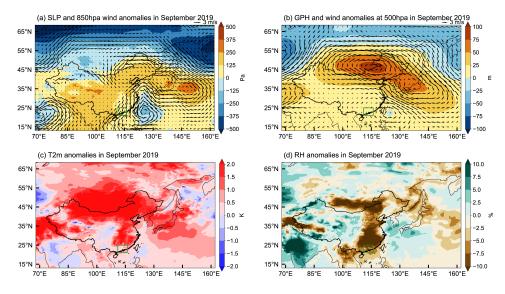
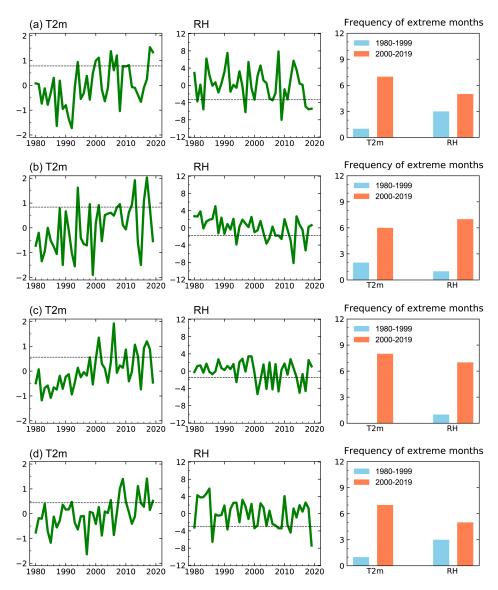


Figure 6. Same as Figure 3 but for the monthly anomalies in September 2019. The green boxesmark PRD.





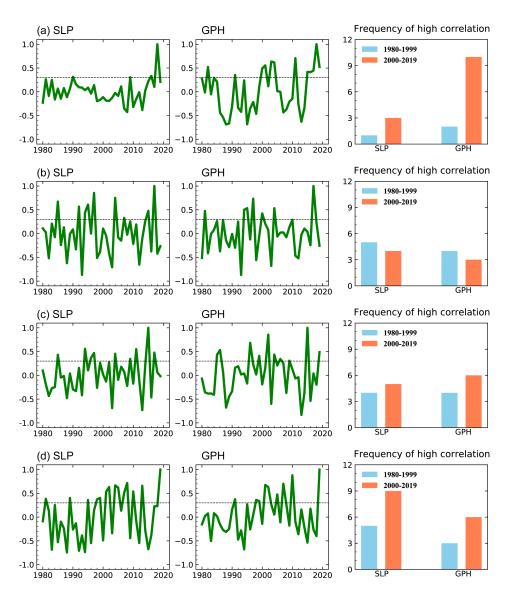




**Figure 7.** Time series of anomalies of T2m (K, left) and RH at 1000 hPa (%, middle) over (a) NCP (115°–120°E, 38°–44°N), (b) YRD ( $120^{\circ}-125^{\circ}E$ ,  $28^{\circ}-32^{\circ}N$ ), (c) SCB ( $102.5^{\circ}-105^{\circ}E$ ,  $30^{\circ}-32^{\circ}N$ ) and (d) PRD ( $110^{\circ}-115^{\circ}E$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





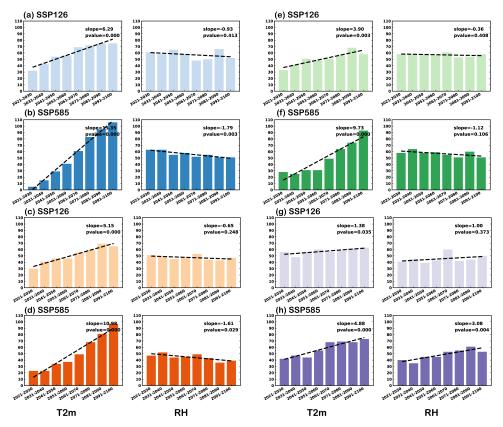


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Figure 8. Time series of spatial correlation in SLP (left) and 500 hPa GPH (middle) anomalies 592 over East Asia and Western Pacific (EAWP, 90°-160°E, 20°-60°N) in June 2018 (a), July 2017 593 594 (b), July 2015 (c) and September 2019 (d) and those of each year during 1980–2019. The dotted lines mark the correlation coefficient of +0.3, which is used as a threshold to define "moderate 595 596 to high correlation". The bar chart (right) represents the frequency of SLP and 500 hPa GPH anomalies in the same months during 1980-1999 (blue) and 2000-2019 (orange) that have 597 moderate to high correlation (>0.3) with those in June 2018, July 2017, July 2015 and 598 September 2019. 599







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





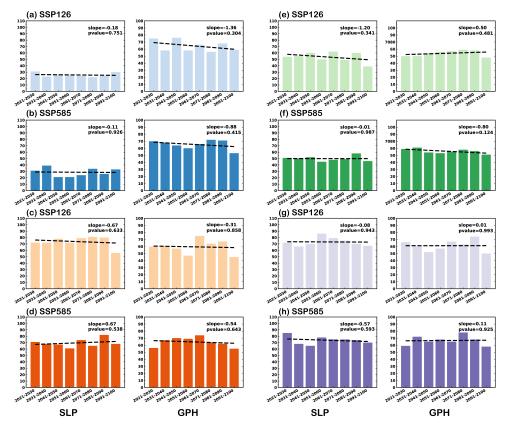


Figure 10. Frequencies of extreme months with SLP and 500 hPa GPH that have moderate to high correlation (>0.3) to those in June 2018 (a, b), July 2017 (c, d), July 2015 (e, f) and September 2019 (g, h) in each 10-year interval during 2021–2100 under two SSPs future scenarios of 13 CMIP6 models. The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