1 Dipole Pattern of Summer Ozone Pollution in the east of China and Its

2 Connection with Climate Variability

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16 Abstract.

17 Surface O₃ pollution has become one of the most severe air pollution problems in China, which makes it of 18 practical importance to understand O₃ variability. A south-north dipole pattern of summer-mean O₃ concentration 19 in the east of China (DP-O₃), which were centered at North China (NC) and the Pearl River Delta (PRD) 20 respectively, has been identified from the simulation of a global 3-D chemical transport model for the period 21 1980-2019. Large-scale anticyclonic (cyclonic) and cyclonic (anticyclonic) anomalies over NC and the PRD 22 resulted in a sharp contrast of meteorological conditions between the above two regions. The enhanced (restrained) 23 photochemistry in NC and restrained (enhanced) O₃ production in the PRD contributed to the DP-O₃. Decreased 24 sea ice anomalies near the Franz Josef Land and associated warm sea surface in May enhanced the Rossby-wave 25 source over northern Europe and West Siberia, which eventually induced an anomalous Eurasia-like pattern to 26 influence the formation of the DP-O₃. The thermodynamic signals of the southern Indian Ocean dipole were stored 27 in the subsurface and influenced spatial pattern of O₃ pollution in the east of China mainly through the Hadley 28 circulation. The physical mechanisms behind the modulation of the atmospheric circulations and related DP-O₃ 29 by these two climate anomalies at different latitudes were evidently verified by large-scale ensemble simulations 30 of the earth system model.

31 Key words: ozone pollution; sea ice; Eurasia pattern; sea surface temperature; meridional circulation

32 1. Introduction

33 Surface O_3 is an important air pollutant. Exposure to high concentrations of O_3 is detrimental to both human 34 health and vegetation ecology (Rider and Carlsten, 2019). Since 2013, surface O₃ concentration has increased 35 over most parts of China, which is largely attributed to changes in anthropogenic emissions (Xu et al. 2018). 36 However, previous studies have shown that in addition to its trend of change, surface O_3 concentration also 37 demonstrated large interannual variations with significant regional differences (Zhou et al. 2013; Chen et al. 2019). 38 Based on analysis of 11 years of observational data over Hong Kong, Zhou et al. (2013) reported that the 39 interannual variation of O₃ concentration observed during 2000–2010 could reach up to 30% of the annual average 40 concentration. The O_3 concentration in Beijing also showed evident interannual variation during 2006–2016. For 41 example, the O₃ concentrations in the summers of 2012–2013 were lower by about 10 ppbv than that in 2011 and 42 2014 (Chen et al. 2019).

43 High O₃ events are usually associated with meteorological factors (e.g., intense solar radiation, high air 44 temperature and low humidity) favorable for O₃ formation, which can accelerate photochemical reaction and 45 weaken the dispersions and depositions (Han et al. 2020). For example, Lu et al. (2019) designed sensitivity 46 simulations to confirm that ozone pollution in China in 2017 was more serious than that in 2016, which was 47 attributed to the large enhancement of nature emissions of ozone precursors caused by hot and dry climate 48 condition in 2017. In the summer of 2013, the Yangtze River Delta experienced a severe heat wave with more 49 stagnant meteorological conditions. The upper-level anticyclonic circulation with sink airflows led to abnormally 50 low atmospheric water vapor content above the Yangtze River Delta and thus less than normal cloud cover, which 51 was conductive to a strong solar radiation environment and significant increases in surface ozone (Pu et al. 2017). 52 On the interannual to decadal time scale, anticyclonic anomalies over North China (NC) were critical for O_3 53 distribution in the summer and remotely linked with the effects of Eurasia teleconnection (EU) and west Pacific 54 patterns (Yin et al. 2019).

The Arctic sea ice (SI) declined rapidly while its variability has been increasing over the past decades, which significantly affected summer atmospheric circulations over Eurasia (Lin and Li 2018). The preceding Arctic SI anomalies could aggravate anomalously high air temperature and drought disasters in NC by triggering EU-like atmospheric responses in summer (Wang and He 2015). Spring SI anomalies in the Barents Sea could prompt the Silk Road Pattern and resulted in a north-south dipole pattern of summer air temperature anomalies in the east of China (Li et al. 2021). When greater than normal SI occurred in the Barents Sea, local 500 hPa geopotential height would decrease and a wave-chain would form, which subsequently induced more precipitation in the south of East 62 China but less precipitation in the north (Wang and Guo 2004). Sea surface temperature (SST) in the Pacific and 63 Indian oceans also have significant effects on atmospheric circulation over the east of China (Li and Xiao 2021; 64 Xia et al. 2021). SST anomalies in the South China Sea and the equatorial Eastern Indian Ocean could trigger the East Asian - Pacific pattern and resulted in a dipole pattern of summer temperature and precipitation in the east 65 66 of China, i.e., areas to the north of the Yangtze River became cold and wet, while areas to the south were hot and 67 dry (Han and Zhang 2009; Li et al. 2018). Tian and Fan (2019) found that winter SST in the southern Indian 68 Ocean might affect spring-summer SST anomalies near Australia. In summer, the anomalous Hadley circulation 69 in the western North Pacific played an important role in summer precipitation over the middle and lower reaches 70 of the Yangtze River.

71 Although great attention in previous studies has been paid to the increase of ozone pollution, little is known about changes in the spatial pattern of summer-mean O₃ in the east of China. As revealed by Yin and Ma (2020), 72 73 the dominant pattern of daily-varying ozone pollution in the east of China showed an interannual variation that 74 was mainly driven by the large-scale western Pacific subtropical high and the East Asian deep trough. For example, 75 the frequent movements of the western Pacific subtropical high and the East Asian deep trough both contributed 76 to the out-of-phase variations in O_3 over North China and the Yangtze River Delta (Zhao and Wang 2017; Yin 77 and Ma 2020). However, to the best of our knowledge, whether the north-south dipole pattern of the summer mean 78 O₃ pollution existed in the east of China still remains unclear. In this study, we attempted to explore the dominant 79 pattern of summertime O₃ in the east of China and associated physical mechanisms behind. Its connections with 80 preceding climate variability were also examined. The remainder of this paper was organized as follows. The data 81 and methods are described in Section 2. Section 3 examined the dipole pattern of summertime O₃ in the east of 82 China and its possible influencing factors. The associated physical mechanisms were studied in Section 4. Major 83 conclusions and discussion are provided in Section 5.

84 **2.** Datasets and methods

85 2.1 Observations and Reanalysis Dataset

Hourly ozone concentration observations from 2015 to 2019 were publicly available at
https://quotsoft.net/air/ and the last accessible data were for 23 September 2020. The relevant data were detrended
before all computations were conducted for the study period.

The meteorological fields data with a horizontal resolution of 0.5° latitude by 0.625° longitude for the period
1980–2019 were taken from the MERRA-2 dataset (Gelaro et al., 2017), including geopotential height at 500 hPa

91 (Z500), surface incoming shortwave flux (Ssr), low and medium cloud cover (Mlcc), precipitation (Prec), 10-m 92 zonal and meridional winds (UV10m), and surface air temperature (SAT) and zonal and meridional winds and 93 vertical velocity at different vertical levels. Monthly Outgoing Longwave Radiation (OLR) data $(1^{\circ} \times 1^{\circ})$ could be acquired from the University of Maryland OLR Climate Data Record portal (http://olr.umd.edu/). Monthly SI 94 95 concentrations and SST ($1^{\circ} \times 1^{\circ}$) for the period 1980 - 2019 were downloaded from the website of the Met Office 96 Hadley Centre (Rayner et al. 2003). Monthly mean subsurface ocean temperatures in the upper 250 m with a 97 horizontal resolution of $1^{\circ} \times 1^{\circ}$ were obtained from the Met Office Hadley Centre EN4 version 2.1 (Good et al. 98 2013).

99 The wave activity flux (WAF) was computed to illustrate the propagation of Rossby wave activities (Takaya100 and Nakamura 2001):

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$$W = \frac{1}{2|\overline{U}|} \left[\frac{\overline{u}(\psi'_x^2 - \psi'\psi'_{xx}) + \overline{v}(\psi'_x\psi'_y - \psi'\psi'_{xy})}{\overline{u}(\psi'_x\psi'_y - \psi'\psi'_{xy}) + \overline{v}(\psi'_y^2 - \psi'\psi'_{yy})} \right]$$

102 where subscripts denote partial derivatives; the overbar and prime represent the climatological mean and 103 anomaly, respectively; ψ' represents the stream function anomaly. U is the horizontal wind speed; u and v are the 104 zonal and meridional wind components, respectively; and W denotes the two-dimensional Rossby WAF. The 105 Rossby wave source $-\nabla \cdot V_x(f + \xi)$ proposed by Sardeshmukh and Hoskins (1988) is also calculated in this 106 study. V, ξ and f refer to the horizontal wind velocity, relative vorticity and geostrophic parameter, 107 respectively. ∇ is horizontal gradient; subscript χ represents divergent component.

108 2.2 1980–2019 O₃ concentrations simulated by GEOS-Chem

Hourly ozone concentrations were simulated by the nested-grid version of the global 3-D chemical transport model (GEOS-Chem), which included detailed description of oxidant–aerosol chemistry. The model was driven by MERRA-2 assimilated meteorological data (Gelaro et al. 2017). The nested grid over China (15–55°N, 75– 135°E) had a horizontal resolution of 0.5° latitude by 0.625° longitude and consisted of 47 vertical layers up to 0.01 hPa. The GEOS-Chem model included the fully coupled O_3 –NOx–hydrocarbon and aerosol chemistry modules with more than 80 species and 300 reactions (Bey et al. 2001).

115 Chemical and physical processes were examined using the outputs of GEOS-Chem. Because non-local 116 planetary boundary layer (PBL) mixing was used, emissions and dry deposition trends within the PBL were 117 applied within the mixing (Holtslag and Boville, 1993). Compared with other terms, the value of wet deposition 118 was extremely small, so it was not considered in this study (Liao et al., 2006). Consequently, the major chemical 119 and physical processes related to meteorological conditions included the chemistry, convection, PBL mixing, 121 The GEOS-Chem model has been widely used to examine historical O₃ changes in China. Yang et al. (2014) 122 evaluated the simulated interannual variation of June-July-August (JJA) surface-layer O₃ concentration at the Hok Tsui station (22°13'N, 114°15'E). They found that the model could well capture the peaks and troughs of the 123 observed JJA O_3 concentration with a high correlation coefficient of +0.87 (exceed the 99% confidence level) 124 125 between simulations and observations. Moreover, the model could also realistically simulate the spatial distribution of O₃, and the spatial correlation coefficient between simulations and observations in the summer of 126 127 2017 could reach up to 0.89 (Li et al. 2019). These studies indicated that the GEOS-Chem model could capture 128 the interannual variation and distribution of the surface O3 concentration fairly well.

129 The GEOS-Chem model successfully reproduced the dominant patterns of summer O₃ pollution on a daily 130 scale from 2015 to 2019 (Yin and Ma 2020). In this study, we first simulated the maximum daily average 8 h 131 concentration of O₃ (MDA8 O₃) from 2015 to 2019 and evaluated the performance of GEOS-Chem. The simulated 132 spatial distribution of MDA8 O_3 was similar to that of observations with a spatial correlation coefficient of 0.87 133 (Fig. 1a). Compared the simulated and observed summer mean MDA8 O₃ concentrations in NC and the PRD, which had a low bias with a mean absolute error of 5.7 μ g m⁻³ and 12.1 μ g m⁻³ in the PRD and NC, respectively. 134 135 The values of root mean square error / mean were 15.8 % and 8.1 % in NC and the PRD, respectively. The observed 136 and simulated summer MDA8 O3 anomalies in the east of China also presented consistent interannual differences 137 (Fig. S1a, b). The high consistency in both the temporal and spatial distributions between the simulations and 138 observations provided a solid evidence to support the feasibility of the present study.



Figure 1 (a) Spatial distributions of observed (dots) and GEOS-Chem simulated (shading) summer-mean MDA8 O₃ (unit: μg
 m⁻³) for the period 2015–2019. (b) The second EOF spatial pattern of simulated summer-mean MDA8 O₃ from 1980 to 2019.

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- 142 The simulated O₃ concentrations were produced by GEOS-Chem with fixed emissions but changing meteorological conditions
- 143 from 1980 to 2019. The green boxes represent the areas of NC and the PRD.

Based the above results, the GEOS-Chem model was then driven by fixed anthropogenic and natural emissions in 2010 and changing meteorological fields from 1980 to 2019 to highlight the impact of climate variability on O₃ concentration. Results of this simulation were analyzed to reveal the dominant pattern of ozone pollution in the east of China in summer and its relationship with preceding climate anomalies.

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2.3 Numerical experiments with CESM-LE

149 To provide evidences that support the proposed connections between SI and SST and large-scale atmospheric 150 circulations, the simulations of the Community Earth System Model Large Ensemble (CESM-LE) were employed 151 (Kay et al. 2015). The CESM consists of coupled atmosphere, ocean, land, and sea ice component models. The 152 40-member ensemble of CESM-LE simulations over the period (1980-2019) includes a historical simulation 153 (1980–2005) and a representative concentration pathway (RCP) 8.5 forcing simulation (2006–2019). To confirm 154 the impact of preceding climate variability and associated physical mechanisms, composite analyses were 155 conducted based on the three years with the lowest and highest simulated preceding climatic variability for a 156 particular month in each member. The composite results of atmospheric circulations could be considered as the 157 relevant atmospheric responses associated with the preceding climate variability.

158 **3.** Dipole pattern of summer O₃ and possible influencing factors

159 As aforementioned, the GEOS-Chem model has a good performance in simulating O₃ concentration. The 160 summer O₃ concentrations from 1980 to 2019 was simulated by GEOS-Chem, and the EOF approach was applied 161 to the GEOS-Chem simulation to explore the dominant patterns of summer mean O₃ pollution in the east of China. 162 Percentage contributions to the total variance by the first and second EOF modes were 39% and 17.5%, 163 respectively. The significance test of the EOF eigenvalues confirmed that the first and second patterns were 164 distinctly separated (passing the North test, North et al, 1982). The first EOF pattern displayed a monopole pattern 165 (Fig. S2). The second EOF pattern presented a north-south dipole pattern of O₃ (DP-O₃) distribution in the east of 166 China with the two centers located in NC and the Pearl River Delta (PRD, Fig. 1b), respectively. Observations 167 have shown that high O₃ concentration frequently occurs in NC, and O₃ pollution in the PRD has become 168 increasingly serious in recent years (Liu et al. 2020). Furthermore, about 80% of the MDA8 O₃ anomalies in NC 169 were in opposite sign to those in PRD during 2015–2019 (Fig. S1a, b). Therefore, despite the fact that it was only 170 the second leading EOF mode, we still focused on the investigation of $DP-O_3$ in the present study, since it was 171 more similar to the actual pollution situation. Impacts of climate variability are also analyzed.

The MDA8 O₃ anomalies were divided into positive (P) and negative phases (N) of DP-O₃ (Fig. S3). For convenience, DP-O₃P and DP-O₃N were defined by the EOF time series of DP-O₃ greater than 1 standard deviation and less than $-1 \times$ standard deviation, respectively. The DP-O₃P corresponded to positive anomalies of MDA8 O₃ in the north and negative anomalies in the PRD (Fig. S3a). In contrast, high concentration of O₃ occurred in the PRD and low concentration center appeared in NC under the DP-O₃N condition (Fig. S3b). The correlation coefficient between time series of DP-O₃ and MDA8 O₃ difference between NC and the PRD was 0.91, indicating that DP-O₃ reflected the opposite changes of O₃ concentration in NC and the PRD.

179 With fixed emissions, the changes in O_3 concentrations from 1980 to 2019 were solely caused by 180 meteorological conditions. The time series of DP-O₃ showed a strong interannual variation (Fig. 2). Composite 181 differences in large-scale atmospheric circulation and meteorological condition related to DP-O₃ between the 182 positive and negative phases (DP-O₃P minus DP-O₃N) were analyzed to explore the impacts of atmospheric 183 circulation on photochemical reactions and accumulations of various pollutants in the above two areas. During the positive phase of DP-O₃, cyclonic and anticyclonic anomalies in the middle troposphere were found over the 184 185 PRD and NC (CPRD and ACNC) (Fig. 3a), respectively. The CPRD and accompanied southerly winds in the PRD 186 efficiently transported clean and moist air from the sea to the PRD (Fig. 3c). Furthermore, low and medium cloud 187 covers were significantly increased, which led to weak solar radiation and reduced photochemical reactions (Fig. 188 3b). A moist, cool environment and weak solar radiation were conductive to low O₃ concentration in the PRD. On 189 the other hand, the positive anomalies of geopotential height in NC increased surface air temperature (Fig. 3a), 190 resulting in a dry environment with decreased cloud covers and sunny weather (Fig. 3b, c).

191 In order to provide a more quantitative evaluation of the contribution of chemical and physical processes, in 192 Fig. 3d, we examine the area-averaged differences in O_3 changes for NC and PRD. Chemistry represents the 193 changes in net chemical production, which appears to be the dominating process, leading to the greatest O_3 change 194 between NC and the PRD (12.3 Tons d⁻¹, Fig. 3d). Transport represents the change in horizontal and vertical 195 advection of ozone. Depending on the ozone concentration gradient and wind anomalies, the transport difference between NC and PRD is 3.1 Tons d⁻¹ (Fig. 3d). Convection changes slightly in NC and PRD. As the mixing 196 197 process transports ozone along the vertical concentration gradient, it generally contributes negatively to the total 198 ozone change. The above analysis indicates that different meteorological conditions between NC and the PRD led 199 to the difference of O_3 concentration in the two regions (differed by 5.2 Tons d⁻¹), which eventually contributed 200 the formation of DP-O₃.



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Figure 2 Variations in standardized DP-O₃ time series (black), May SI near the Franz Josef Land (SI_{FIL}, red), January– February–March mean Subtropical Indian Ocean Dipole (SIOD, blue), and SEI (green) from 1980 to 2019. SEI defined as the weighted average of SI_{FIL} and SIOD. The correlation coefficients of the DP-O₃ with SI_{FIL} (red), SIOD (blue), and SEI (green) were shown in the figure.



207 Figure 3 Composite summer atmospheric circulations associated with the DP-O3 (DP-O3P minus DP-O3N) for the period 1980 208 to 2019, including (a) surface air temperature (SAT, unit: K, shadings) and geopotential height at 500 hPa (unit: 10 gpm, 209 contours), (b) surface incoming shortwave flux (Ssr, unit: W m⁻², shadings) and low and medium cloud cover (Mlcc, unit: 1, 210 contours), and (c) precipitation (Prec, unit: mm, shadings) and surface wind (unit: m s⁻¹, arrows). The white dots indicate that 211 the composites with shading were above the 90% confidence level. The black boxes in (a) indicate the centers of the AC_{NC} and 212 CPRD, respectively. The green boxes in (b) and (c) represent the areas of NC and the PRD. Composites of the summer mass 213 fluxes of O₃ (d) associated with the DP-O₃ (DP-O₃P minus DP-O₃N) for the area-averaged differences (NC minus PRD) from 214 1980 to 2019. The bottom axis gives the names of the chemical and physical processes: chemical reaction (Chem), convection 215 (Conv), PBL mixing (Mix), transport (Trans) and their sum (Sum).

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216 Arctic SI in May was closely related to summer O₃ pollution in NC (Yin et al. 2019), but its effects on the 217 north-south dipole distribution of O₃ had not been studied. The meridional O₃ dipole pattern in the east of China 218 was positively correlated with SI anomalies near the Franz Josef Land (SIFIL). Note that the correlation between 219 them remains unchanged after the signal of El Niño-Southern Oscillation (ENSO) was removed. The area-220 averaged (82-88°N, 3°W-60°E; 79-88°N, 60-90°E; denoted by the green boxes in Fig. 4a) SI in May was 221 calculated and defined as the SIFIL index, whose linear correlation coefficient with the time series of DP-O3 was 222 0.52 (exceeding the 99% confidence level). When the SI_{FIL} anomalies were significant (i.e., |anomalies| > its one 223 standard deviation), the occurrence probability of the DP-O₃ in the same phase was 83% (Fig. 2). Furthermore, 224 the active centers of the anomalous atmospheric circulations and meteorological conditions associated with SI_{FIL} 225 in the east of China were similar to that of the DP-O₃ (i.e., NC and PRD). That is, positive SI_{FIL} anomalies were 226 conductive to less (more) precipitation, less (more) cloud cover, and strong (weak) solar radiation in NC (PRD)

227 (Fig. 4c, Fig. S4). The chemical and physical processes of ozone production in GEOS-Chem simulations were 228 analyzed. The difference of chemical reactions between NC and PRD had a large positive value (11.6 Tons d^{-1}), 229 and the difference of the sum of all chemical and physical processes was 7.0 Tons d^{-1} (Fig. 4c), resulting in DP-230 O₃.



232 Figure 4 Composites of (a) May SI concentration and (b) JFM SST associated with the DP-O₃ (DP-O₃P minus DP-O₃N) from 233 1980 to 2019. The green boxes in (a) and (b) indicate where the SIFIL and SIOD indices are calculated, respectively. The white 234 dots indicate that the composites were above the 90% confidence level. Composite summer meteorological conditions, 235 circulations and mass fluxes of O3 associated with (c) SIFJL (positive SIFJL years minus negative SIFJL years) and (d) SIOD 236 (positive SIOD years minus negative SIOD years) from 1980 to 2019. The bottom axis gives the names of the meteorological 237 conditions and chemical and physical processes: the differences between AC_{NC} and C_{PRD} (unit: 10 gpm), surface incoming 238 shortwave flux (Ssr, unit: W m⁻²), surface air temperature (SAT, unit: K), and precipitation (Prec, unit: mm); chemical reaction 239 (Chem, unit: Tons d⁻¹), convection (Conv, unit: Tons d⁻¹), PBL mixing (Mix, unit: Tons d⁻¹), transport (Trans, unit: Tons d⁻¹) 240 and their sum (Sum, unit: Tons d^{-1}).

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In addition to the signal from the Arctic, SST as an effective external forcing also has significant influences on summer climate in the east of China (Li et al. 2018). Therefore, it was important to answer the question whether SST could affect the DP-O₃ in the east of China in summer. Large anomalies of preceding January–February– March (JFM) SST over the southern Indian Ocean was obvious when we evaluated the relationship between the DP-O₃ and previous SST. After removing the influence of ENSO, the SST signal in the southern Indian Ocean still maintains (Fig. 4b). The two regions with significant anomalies were similar to the Subtropical Indian Ocean 247 Dipole (SIOD) regions found by Behera and Yamagata (2001). Variance analysis and correlation analysis of SST 248 in the Indian Ocean also indicated that a SST dipole type oscillation occurred in the southern Indian Ocean, which 249 usually developed in the preceding winter and reaches its strongest in the subsequent January to March (Jia and 250 Li 2013). The difference between the mean SST of the two regions (29–40°S, 31–62°E and 7–25°S, 55–86°E; 251 green box in Fig. 4b; the southwest positive pole minus the northeast negative pole) was defined as the SIOD 252 index and calculated (Fig. 2). The linear correlation coefficient between the SIOD index and the time series of 253 DP-O₃ from 1980 to 2019 was 0.44 (significant at the 99% confidence level). When the SIOD anomalies were 254 significant (i.e., |anomalies| > its one standard deviation), the occurrence probability of DP-O₃ in the same phase 255 is 82% (Fig. 2). Furthermore, the composite meteorological conditions in the positive and negative phases of 256 SIOD had similar centers to that of DP-O₃. That is, the anticyclone over NC was always accompanied by hot-dry 257 meteorological condition, while the cyclone over PRD was always accompanied by cool-moist environment (Fig. 258 4d; Fig. S5). The chemical reactions increased 12.3 Tons d^{-1} in NC comparing to those in the PRD (Fig. 4 d), 259 indicating that the strong solar radiation and high temperature conditions actually enhanced the chemical reactions 260 in the atmosphere to produce more O_3 in NC.

261 4. Associated physical mechanisms

262 Changes in SIFIL and SIOD both could possibly contribute to the formation of DP-O3. Note that SIFIL and SIOD have few years of common significant anomalies, more than 78% of the individual sample years were used 263 264 to make composite with both indices. The correlation coefficient between them was only 0.21 and was not 265 significant, indicating that SIFJL and SIOD were independent of each other. Several previous studies have 266 documented that the preceding Arctic SI anomalies could trigger EU-like atmospheric responses in the subsequent 267 summer, and thus influenced the climate in the east of China (Wang and He 2015). Corresponding to reduced 268 SI_{FJL}, SST anomalies in the Barents and Kara Sea were significantly positive and gradually increase from May to 269 summer months (Fig. 5a, b). The warm SST anomalies influenced local heat anomalies and caused anomalous 270 atmospheric circulations. Following the decrease in SIFIL, anomalous divergent winds appeared in the mid-271 troposphere, which were accompanied by warm SST anomalies and negative velocity potential anomalies (yellow 272 box in Fig. 5c). As proposed by Xu et al., (2021), the rotational component of the anomalous divergent winds 273 could spread to the south and force the vorticity generation over Eurasia. Thus, during the subsequent summer, 274 significant convergence and positive velocity potential with a positive Rossby wave source anomaly occured over 275 northern Europe and West Siberia (green box in Fig. 5d). We also used the SST anomalies associated with SI_{FJL} 276 (in Barents and Kara Sea in JJA) to composite relevant variables. Significant convergence, positive velocity 277 potential, and positive Rossby source anomaly all appeared over Europe and West Siberia in JJA (Fig. S6). This

278 indicated that positive anomalies of Rossby-wave source over Europe and West Siberia could be generated

279 by local heat anomalies associated with decreased SI_{FJL} in the Barents and Kara Sea.



Figure 5 Composites of (a) May Arctic SST (unit: K), (c) velocity potential (unit: $10^5 \text{ m}^2 \text{ s}^{-1}$, shading) and divergent wind at 500 hPa (unit: m s⁻¹, arrows), and (e) Rossby wave source anomalies at 500 hPa (unit: 10^{-11} s^{-2}) associated with SI_{FJL} index (negative SI_{FJL} years minus positive SI_{FJL} years) from 1980 to 2019. The back box in (a) and (b), yellow box in (c) and (e) and green box in (d) and (f) represents the center of the SST, velocity potential and Rossby wave source anomaly associated with SI_{FJL}, respectively. The white dots indicate that the composites with shading were above the 90% confidence level.

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Moreover, corresponding to the decreased SI_{FJL}, the anomalous Rossby WAF propagated from Europe and West Siberia (consistent with the aforementioned Rossby wave source) to Northeast China and enhanced the cyclonic anomaly nearby (Fig. 6a). The anomalous cyclonic circulation caused ascending motion from the surface





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Figure 6 Composites of (a) wave activity flux anomalies (unit: $m^2 s^{-2}$, arrows), geopotential height (unit: gpm, shading) at 500 hPa and (b) mean wind (unit: $m s^{-1}$, arrows), omega (unit: 10^{-2} Pa s⁻¹, shading) over $100-130^{\circ}$ E, and the anomalies of AC_{NC} and C_{PRD} (unit: gpm, bar) in summer associated with SI_{FJL} index (negative SI_{FJL} years minus positive SI_{FJL} years) from 1980 to 2019. The green boxes in (a) represent the centers of the EU-like pattern. The white dots indicate that the composites with shading were above the 90% confidence level.

The relationship between the preceding May SI anomalies and the JJA EU-like pattern was also confirmed by large ensemble simulations of CESM during 1980–2019. According to the simulated sea ice fraction near the Franz Josef Land, the three years with the lowest and highest SI in each member were selected to construct the 306 composite maps based on all the 40 available members. The difference in JJA geopotential height at 500 hPa 307 represented the atmospheric response to declining May SI_{FJL}. As shown in Fig. 7, the decline of SI_{FJL} in May led 308 to an EU-like pattern in the subsequent summer over Eurasia, which was in good accordance with the observed 309 result (Fig. 6a). The anticyclonic and cyclonic anomalies shown in the geopotential height at 500 hPa (i.e., AC_{NC} 310 and C_{PRD}) in summer were also well reproduced by over 60% of the members. The above results confirmed the 311 robustness of the physical mechanisms proposed in the present study.



Figure 7 Composite differences of geopotential height at 500 hPa in JJA between three low and high SI_{FIL} years based on the ensemble of 40 CESM-LE simulations during 1980–2019. The black dots indicate that the mathematical sign of the composite results of more than 60 % of the members is consistent with the ensemble mean. The black boxes represent the centers of the EU-like pattern.

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317 SIOD could influence atmospheric anomalies and distribution of summer precipitation in China mainly 318 through Hadley circulation (Liu et al. 2019). Can SIOD anomalies also influenced the DP-O₃ via meridional 319 atmospheric forcing? Despite the significant correlation between SIOD anomalies (defined by SST) and the DP-O₃ in the east of China (Fig. 4b), it should be noted that the thermodynamic signals in the southern Indian Ocean 320 not only existed on the sea surface but also extended to the subsurface (Fig. S7). As time goes by, the center of 321 negative SST anomalies moved to the northeast possibly due to the eastward movement of atmospheric forcing 322 323 caused by the mean westerly flow (Behera and Yamagata 2001). When it moved to the vicinity of Sumatra Island 324 in JJA, the abnormally cold signals of SST could extend downward from the surface to 60m (black box in Fig. 325 8a). The area-averaged (black box in Fig. 8a) summer-mean subsurface ocean temperature of 0-60m was defined 326 as the SOT index and calculated. Affected by negative SOT anomalies near Sumatra Island, the equatorial eastern 327 Indian Ocean convection was suppressed (indicated by positive anomalies of OLR in Fig. 8b) and significant 328 divergence prevailed in the lower troposphere (Fig. 8c). As a result, anomalous downward air flow developed near Sumatra Island from 300 hPa to the surface (about 20-5°S in Fig. 8d). This anomalous downward air flow 329 330 modulated the meridional circulation over 90-120 °E by strengthening the abnormal upward airflow at 20°N and

- downward airflow at 30°N. Thus, the AC_{NC} and C_{PRD} were enhanced simultaneously (Fig. 8d). Overall, following
- the positive phase of SIOD, the cold signal of SOT anomalies changed the meridional circulation in the subsequent JJA and strengthened the C_{PRD} and AC_{NC} in the troposphere above the east of China. Under these large-scale atmospheric anomalies, O_3 concentrations became higher in NC, whereas the generation of surface O_3 were
- 335 weakened in the PRD.



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337 Figure 8 (a) Composites of mean 0-60m subsurface ocean temperature (unit: K) in summer associated with the SIOD (positive 338 SIOD years minus negative SIOD years) from 1980 to 2019. The green boxes represent the centers of the SIOD, and the black 339 box indicates where the SOT index is calculated. Composites of (b) OLR (unit: W m⁻²) and (c) velocity potential (unit: 10⁵ m² 340 s^{-1} , shadings) and divergent winds (unit: m s^{-1} , vectors) at 10 m in summer associated with SOT indexes of opposite sign 341 (negative SOT years minus positive SOT years). The black box represents the center of the SOT. (d) Composites of summer 342 mean winds (unit: m s⁻¹, arrows) and omega (unit: 10⁻² Pa s⁻¹, shadings) over 90–120°E, and the anomalies of AC_{NC} and C_{PRD} 343 (unit: gpm, bars) associated with SOT indexes of opposite sign. The white dots indicate that the composites with shading were 344 above the 90% confidence level.

The CESM-LE datasets were also used to verify the statistical correlation between the preceding SIOD and large-scale atmospheric circulations in JJA. The composite differences of SIOD in JFM between the three high years and three low years of SST simulated by each ensemble member during 1980–2019 were investigated based on the ensemble of 40 CESM-LE simulations. The composite results (positive SIOD years minus negative SIOD years) of atmospheric circulations could be considered as the relevant atmospheric circulation responses associated with differences in SIOD. More than 60% of the CESM ensemble members could well reproduce the

- anticyclonic circulation over NC and the cyclonic circulation over the PRD in summer at 500hPa (Fig. 9). That is,
- 352 the CESM-LE also confirmed the relationship between the previous JFM SIOD anomaly and the DP-O₃-related
- 353 atmospheric circulations (i.e., AC_{NC} and C_{PRD}) in subsequent JJA.



354

Figure 9 Composite differences of geopotential height at 500 hPa in JJA between three high and low SIOD years based on the ensemble of 40 CESM-LE simulations during 1980–2019. The black dots indicate that the mathematical sign of the composite results of more than 60 % of the members is consistent with the ensemble mean. The black boxes represent the centers of AC_{NC} and C_{PRD}, respectively.

359 5. Conclusions and discussions

360 In general, the O_3 concentrations in NC were substantially high and the problem of O_3 pollution in the PRD 361 has become increasingly prominent in recent years. A south-north dipole pattern of O₃ concentration in the east of 362 China was identified based on GEOS-Chem simulations with fixed emissions and changing meteorological condition from 1980 to 2019. The DP-O₃ pattern presented opposite centers in NC and PRD. Corresponding to 363 364 the positive phase of DP-O₃, cyclonic and anticyclonic anomalies were located over the PRD and NC respectively, 365 which resulted in dry and hot climate in NC, while the environment in the PRD region was cool and moist. The 366 opposite was true in the negative phase of DP-O₃. During positive phases, the meteorological condition mentioned 367 above significantly enhanced photochemical reactions in NC but suppressed O₃ production in the PRD, and thus 368 make great contributions to the south-north dipole pattern of O₃ in the east of China.

Arctic SI near the Franz Josef Land in May played an important role in the occurrence of DP-O₃. The warm SST anomalies associated with less SI_{FJL} could induce divergent wind field and vorticity advection in the upper layer, and enhanced positive Rossby wave source over northern Europe and West Siberia in summer. An EU-like 372 pattern was triggered in Eurasia (solid lines in Fig. 10), which could enhance the DP-O₃-related atmospheric 373 circulation (i.e., AC_{NC} and C_{PRD}) in JJA. As a result, meteorological conditions for O₃ concentration were 374 completely different between NC and PRD, which eventually contributed the formation of DP-O₃. In addition, the 375 precursory climatic driving signal of SIOD anomalies in the low latitudes in JFM was also closely linked to DP-O3. The thermodynamic signal of SIOD could be stored in the subsurface, and the center of negative SST 376 377 anomalies moved to the vicinity of Sumatra Island in summer. The meridional circulation intensified in summer 378 (dashed lines in Fig. 10), which, along with the enhancement of the AC_{NC} and C_{PRD} over the east of China, 379 effectively increased O₃ concentration in NC but suppressed the generation of surface O₃ in the PRD. The linkages 380 and corresponding physical mechanisms were well reproduced by the large CESM-LE ensemble simulation.



Figure 10 Schematic diagrams of the associated physical mechanisms. The May SI anomalies near the Franz Josef Land (red shadings) could trigger an EU-like pattern in the atmosphere in summer, which enhances the anticyclonic anomaly over NC and the cyclonic anomaly over the PRD. The thermodynamic signal of the preceding SIOD (contours) could be stored in the subsurface and the center of negative SST anomalies moves to the vicinity of Sumatra Island in summer (blue shading). The meridional circulation was enhanced in summer (dashed lines), along with the enhancement of AC_{NC} and C_{PRD} over eastern China. The solid lines indicate the anomalous atmospheric circulations affected by SI_{F/L}, while the dashed lines indicate the anomalous atmospheric circulations affected by SIOD.

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389 The above analysis has revealed that the DP-O₃ is independently affected by SIOD and SI_{FJL} from 1980 to 390 2019. We attempted to discuss the combined impacts of the two precursory climatic drivers in the present 391 study. For this purpose, a synthetic climate variability index SEI, defined as the weighted average of SI_{FJL} and 392 SIOD, is calculated by

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$$SEI = \frac{r_1 \times SI_{FJL} + r_2 \times SIOD}{|r_1| + |r_2|}$$

394 where r_1 and r_2 were the correlation coefficients of SI_{FJL} ($r_1 = 0.52$) and SIOD ($r_2 = 0.44$) with the DP-O₃ time series, respectively. The correlation coefficient between SEI and DP-O₃ was 0.62 (Fig. 2, exceeding the 395 396 99% confidence level). When the SEI anomalies were significant, the occurrence probability of the DP-O₃ in the 397 same phase was 93% (Fig. 2), which is higher than that based on individual influences of the two factors. 398 Composite atmospheric circulation analysis has been carried out based on years of positive and negative SEI 399 anomalies, and the results are shown in Fig. 11a. The composite atmospheric circulation based on the SEI index 400 was stronger, resulting in the concentrations of MDA8 O₃ in NC was 11.74 µg m⁻³ higher than that in PRD (Fig. 401 11b). The main areas influenced by SI and SST were slightly different. Although the two precursory climatic 402 drivers both could affect the atmospheric circulations over NC and the PRD, SI_{FIL} mainly affected atmospheric 403 circulation anomaly over NC, while SIOD played a major role in the PRD. However, climate variabilities at 404 different latitudes jointly facilitated the dipole pattern of O₃ in the east of China from 1980 to 2019.



Figure 11 (a) Composites of geopotential height at 500 hPa (unit: gpm, shadings) in summer associated with the SEI (positive SEI years minus negative SEI years) from 1980 to 2019. The red and blue lines indicate areas where the composite geopotential height anomalies associated with SI_{FJL} and SIOD exceed the 90% confidence level, respectively. The black boxes represent the centers of AC_{NC} and C_{PRD}, respectively. (b) Composite differences of the detrended summer-mean MDA8 O₃ (unit: μ g m⁻³) simulated by GEOS-Chem model between high and low SEI years during 1980–2019. The white dots indicate that the composite differences are above the 90% confidence level. The green boxes represent the areas of NC and the PRD.

412 The north-south dipole pattern of O_3 in the east of China in summer and its relationship with climate factors 413 were clearly revealed in this study, yet some questions still remain unanswered and should be investigated in the 414 future. The GEOS-Chem model simulations were used to explore the dominant pattern of O_3 in the east of China in summer due to the short sequence of O_3 observations. Although the GEOS-Chem demonstrated a good performance based on evaluation, there still exist some differences between the simulations and observations. In addition, statistical and numerical methods were used to reveal and verify the physical mechanisms behind the dipole pattern of O_3 in the east of China and its relation with climate variability. However, further numerical experiments should be carried out in the future. For example, coupled climate-chemistry models should be used to not only simulated the influence of climate driving factors on O_3 pattern, but also revealed the effect of individual climate factors as well as their comprehensive effects.

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423 Data Availability.

Hourly O₃ concentration data could be downloaded from https://quotsoft.net/air/ (Ministry of Environmental
Protection of China, 2020). Sea ice concentration, sea surface temperature, and subsurface ocean temperature data
were from https://www.metoffice.gov.uk/hadobs/ (Met Office Hadley Centre, 2021). Monthly-mean MERRA-2
reanalysis dataset was available at https://disc.gsfc.nasa.gov/datasets?page=1 (MERRA-2, 2021). The monthly
OLR data could be acquired from http://olr.umd.edu/ (University of Maryland OLR Climate Data Record portal,
2021).

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431 Authors' contribution

432 Yin Z. C. designed the research. Ma X. Q. did the statistical analysis and implemented the GEOS-Chem
433 simulations. Yin Z. C. and Ma X. Q. prepared the manuscript.

434

435 Competing interests

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

437

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558 Table and Figure captions

- 559 Figure 1 (a) Spatial distributions of observed (dots) and GEOS-Chem simulated (shading) summer-mean MDA8
- 560 O_3 (unit: $\mu g m^{-3}$) for the period 2015–2019. (b) The second EOF spatial pattern of simulated summer-mean MDA8
- 561 O₃ from 1980 to 2019. The simulated O₃ concentrations were produced by GEOS-Chem with fixed emissions but
- changing meteorological conditions from 1980 to 2019. The green boxes represent the areas of NC and the PRD.
- 563 Figure 2 Variations in standardized DP-O₃ time series (black), May SI near the Franz Josef Land (SI_{FJL}, red),
- January-February-March mean Subtropical Indian Ocean Dipole (SIOD, blue), and SEI (green) from 1980 to
- 565 2019. SEI defined as the weighted average of SI_{FJL} and SIOD. The correlation coefficients of the DP-O₃ with SI_{FJL}
- 566 (red), SIOD (blue), and SEI (green) were shown in the figure.
- 567 Figure 3 Composite summer atmospheric circulations associated with the DP-O₃ (DP-O₃P minus DP-O₃N) for 568 the period 1980 to 2019, including (a) surface air temperature (SAT, unit: K, shadings) and geopotential height at 569 500 hPa (unit: 10 gpm, contours), (b) surface incoming shortwave flux (Ssr, unit: W m⁻², shadings) and low and 570 medium cloud cover (Mlcc, unit: 1, contours), and (c) precipitation (Prec, unit: mm, shadings) and surface wind 571 (unit: m s⁻¹, arrows). The white dots indicate that the composites with shading were above the 90% confidence 572 level. The black boxes in (a) indicate the centers of the AC_{NC} and C_{PRD} , respectively. The green boxes in (b) and 573 (c) represent the areas of NC and the PRD. Composites of the summer mass fluxes of O_3 (d) associated with the 574 DP-O₃ (DP-O₃P minus DP-O₃N) for the area-averaged differences (NC minus PRD) from 1980 to 2019. The 575 bottom axis gives the names of the chemical and physical processes: chemical reaction (Chem), convection (Conv),
- 576 PBL mixing (Mix), transport (Trans) and their sum (Sum).
- 577 **Figure 4** Composites of (a) May SI concentration and (b) JFM SST associated with the DP-O₃ (DP-O₃P minus 578 DP-O₃N) from 1980 to 2019. The green boxes in (a) and (b) indicate where the SI_{FJL} and SIOD indices are 579 calculated, respectively. The white dots indicate that the composites were above the 90% confidence level. 580 Composite summer meteorological conditions, circulations and mass fluxes of O₃ associated with (c) SI_{FJL}
- 581 (positive SI_{FJL} years minus negative SI_{FJL} years) and (d) SIOD (positive SIOD years minus negative SIOD years)
- from 1980 to 2019. The bottom axis gives the names of the meteorological conditions and chemical and physical
- 583 processes: the differences between AC_{NC} and C_{PRD} (unit: 10 gpm), surface incoming shortwave flux (Ssr, unit: W
- 584 m⁻²), surface air temperature (SAT, unit: K), and precipitation (Prec, unit: mm); chemical reaction (Chem, unit:
- 585 Tons d^{-1}), convection (Conv, unit: Tons d^{-1}), PBL mixing (Mix, unit: Tons d^{-1}), transport (Trans, unit: Tons d^{-1})
- 586 and their sum (Sum, unit: Tons d^{-1}).

- 587 Figure 5 Composites of (a) May Arctic SST (unit: K), (c) velocity potential (unit: 10⁵ m² s⁻¹, shading) and
- 588 divergent wind at 500 hPa (unit: m s⁻¹, arrows), and (e) Rossby wave source anomalies at 500 hPa (unit: 10^{-11} s⁻²)
- associated with SI_{FJL} index (negative SI_{FJL} years minus positive SI_{FJL} years) from 1980 to 2019. The back box in
- 590 (a) and (b), yellow box in (c) and (e) and green box in (d) and (f) represents the center of the SST, velocity potential
- 591 and Rossby wave source anomaly associated with SI_{FJL}, respectively. The white dots indicate that the composites
- 592 with shading were above the 90% confidence level.
- 593 Figure 6 Composites of (a) wave activity flux anomalies (unit: m² s⁻², arrows), geopotential height (unit: gpm,

594 shading) at 500 hPa and (b) mean wind (unit: m s⁻¹, arrows), omega (unit: 10^{-2} Pa s⁻¹, shading) over $100-130^{\circ}$ E,

and the anomalies of AC_{NC} and C_{PRD} (unit: gpm, bar) in summer associated with SI_{FJL} index (negative SI_{FJL} years

- 596 minus positive SI_{FIL} years) from 1980 to 2019. The green boxes in (a) represent the centers of the EU-like pattern.
- 597 The white dots indicate that the composites with shading were above the 90% confidence level.
- **Figure 7** Composite differences of geopotential height at 500 hPa in JJA between three low and high SI_{FJL} years based on the ensemble of 40 CESM-LE simulations during 1980–2019. The black dots indicate that the mathematical sign of the composite results of more than 60 % of the members is consistent with the ensemble mean. The black boxes represent the centers of the EU-like pattern.
- 602 Figure 8 (a) Composites of mean 0–60m subsurface ocean temperature (unit: K) in summer associated with the
- 603 SIOD (positive SIOD years minus negative SIOD years) from 1980 to 2019. The green boxes represent the centers
- 604 of the SIOD, and the black box indicates where the SOT index is calculated. Composites of (b) OLR (unit: W m⁻²)
- and (c) velocity potential (unit: $10^5 \text{ m}^2 \text{ s}^{-1}$, shadings) and divergent winds (unit: m s^{-1} , vectors) at 10 m in summer
- associated with SOT indexes of opposite sign (negative SOT years minus positive SOT years). The black box
- 607 represents the center of the SOT. (d) Composites of summer mean winds (unit: m s⁻¹, arrows) and omega (unit:
- 10^{-2} Pa s⁻¹, shadings) over 90–120°E, and the anomalies of AC_{NC} and C_{PRD} (unit: gpm, bars) associated with SOT
- indexes of opposite sign. The white dots indicate that the composites with shading were above the 90% confidencelevel.
- Figure 9 Composite differences of geopotential height at 500 hPa in JJA between three high and low SIOD years based on the ensemble of 40 CESM-LE simulations during 1980–2019. The black dots indicate that the mathematical sign of the composite results of more than 60 % of the members is consistent with the ensemble mean. The black boxes represent the centers of AC_{NC} and C_{PRD} , respectively.
- 615 Figure 10 Schematic diagrams of the associated physical mechanisms. The May SI anomalies near the Franz Josef 616 Land (red shadings) could trigger an EU-like pattern in the atmosphere in summer, which enhances the 617 anticyclonic anomaly over NC and the cyclonic anomaly over the PRD. The thermodynamic signal of the preceding SIOD (contours) could be stored in the subsurface and the center of negative SST anomalies moves to 618 619 the vicinity of Sumatra Island in summer (blue shading). The meridional circulation was enhanced in summer 620 (dashed lines), along with the enhancement of AC_{NC} and C_{PRD} over eastern China. The solid lines indicate the 621 anomalous atmospheric circulations affected by SIFIL, while the dashed lines indicate the anomalous atmospheric 622 circulations affected by SIOD.
- Figure 11 (a) Composites of geopotential height at 500 hPa (unit: gpm, shadings) in summer associated with the
 SEI (positive SEI years minus negative SEI years) from 1980 to 2019. The red and blue lines indicate areas where

- 625 the composite geopotential height anomalies associated with SI_{FJL} and SIOD exceed the 90% confidence level,
- 626 respectively. The black boxes represent the centers of AC_{NC} and C_{PRD}, respectively. (b) Composite differences of
- 627 the detrended summer-mean MDA8 O_3 (unit: $\mu g m^{-3}$) simulated by GEOS-Chem model between high and low
- 628 SEI years during 1980–2019. The white dots indicate that the composite differences are above the 90% confidence
- 629 level. The green boxes represent the areas of NC and the PRD.

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