1	Decadal Changes of Connections among <u>late-spring</u> Snow cover in West
2	Siberia, summer Eurasia Teleconnection and O ₃ -related meteorology in
3	North China
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19	Abstract. Severe surface ozone (O ₃) pollution frequently occurred in North China and obviously damages human
20	health and ecosystems. The meteorological conditions effectively modulate the variations in O ₃ pollution. In this
21	study, the interannual relationship between O3-related meteorology and late-spring snow cover in West Siberia
22	was explored, and the reasons of its decadal change were also physically explained. Before mid-1990s, less snow
23	cover could enhance net heat flux and stimulate positive phase of the Eurasia (EU) teleconnection in summer. The
24	positive EU pattern resulted in hot-dry air and intense solar radiation in North China, which could enhance the
25	natural emissions of O3 precursors and photochemical reactions in the atmosphere closely related to high O3

26 concentrations. However, after the mid-1990s, the south edge of the dense snow cover area in West Siberia shifted

27 northward by approximately 2° in latitude and accompanied radiation and heat flux also retreated toward the polar

region. The connections among snow anomalies, EU pattern and surface O₃ became insignificant and thus

29 influenced the stability of the predictability.

30 Key words: Eurasia pattern; ozone pollution; snow cover; sea ice; Arctic

31 **1. Introduction**

32 The Eurasia teleconnection (EU) pattern is a major quasistationary wave train in the Northern Hemisphere 33 (Wallace and Gutzler, 1981; Wang and Zhang, 2015) and effectively linked the climate variability between the polar region and eastern China (Wang and He, 2015). The EU pattern appears in all seasons and consists of centers 34 35 of geopotential height anomalies over polar region, Mongolia and North China, and the Yellow Sea and Japan Sea 36 (Liu et al., 2014). The impacts of the EU pattern on the Eurasian climate have been investigated by many previous 37 studies. The phase and intensity of the EU pattern have important impacts on the East Asia winter monsoon (Lim 38 and Kam, 2016), as well as on the Siberian High (Gong et al., 2001), subtropical jet and East Asian trough (Liu 39 and Chen, 2012). The enhanced winter monsoons resulted in lower temperatures and less precipitation in East 40 China (Yan et al., 2003). Likewise, the EU pattern significantly influenced the dispersion conditions in North 41 China and thus played important roles in local haze pollution (Li et al., 2019). In summer (June-July-August, JJA), 42 the EU pattern influenced the Ural-blocking high and the East Asian trough and thus played important roles in the 43 variability of summer precipitation over China (Zhang et al., 2018). Similarly, severe summer droughts in North 44 China also had close relationships with the largest anomalies of the EU pattern (Wei et al., 2004). For example, 45 the EU-like anomalous atmospheric circulations in summer 2014 resulted in an above-normal East Asian trough 46 and a southward shift of the west Pacific subtropical high. Consequently, North China suffered from its most 47 severe drought during the period of 1979–2014 (Wang and He, 2015). Moreover, the positive phase of the EU 48 pattern in 2016 favored downward motions and weaker convergences of moisture and thus resulted in high air 49 temperatures and a dry atmosphere in North China (Li et al., 2018).

50 High concentrations of ground-level ozone (O₃) are frequently observed together with dry-hot air and intense 51 solar radiation because photochemical reactions are accelerated under such meteorological conditions (Pu et al., 52 2017). The large-scale atmospheric circulations associated with high-O3-related meteorology in North China 53 appeared as the positive phase of the EU pattern (Yin et al., 2019, 2020a). The anomalous anticyclonic circulations 54 over North China, as one active center of the EU pattern, induced significant descending air flows and thus 55 efficient adiabatic heating and intense sunlight (Gong and Liao, 2019). Generally, numerous nitrogen oxides (NOx) 56 and volatile organic compounds (VOCs) are emitted by human activities and natural sources in North China 57 (Zheng et al., 2018). These precursors of O_3 react under high ultraviolet radiation and generate more O_3 (Fix et 58 al., 2018).

59 The variation in the EU pattern and its linkage with surface O₃ in North China were both driven by preceding 60 spring forcings (Zhang et al., 2018; Yin et al., 2019, 2020a). Arctic sea ice anomalies in spring were proven to be 61 closely related to the summer EU teleconnection pattern; these anomalies then influenced rainfall in China (Wu 62 et al., 2009). Summer surface O_3 in North China closely linked to the variability in May sea ice over the Gakkel 63 Ridge (Figure S1) and the bridge in atmosphere was the EU pattern (Yin et al., 2019, 2020a). However, this 64 relationship between sea ice anomalies and EU pattern showed a decadal change from insignificant to significant 65 after the mid-1990s (Yin et al., 2020a). The east-west dipole of spring snow cover anomalies in Eurasia was 66 closely related to the East Asia summer monsoon by stimulating atmospheric responses such as the EU pattern (Yim et al., 2010). When building a seasonal prediction model of surface O_3 -related meteorology, the May snow 67 68 cover in West Siberia was selected as a predictor and effectively increased the predictability (Yin et al., 2020b). 69 However, the physical mechanisms linking O₃ and snow cover are still unclear.

Two open questions are as follows: (1) Have the links between the EU pattern and O_3 -related meteorology in North China changed over the decades? (2) What is the roles of snow cover anomalies on driving the above connection? This study aimed to answer these unrevealed questions and explain the associated physical mechanisms. The remainder of this paper is organized as follows. Section 2 describes the data and methods, and the decadal changes in relationships between climatic factors were analyzed in Section 3. The physical mechanisms driving the changes were proposed and explained in Section 4. The main conclusions and necessary discussion of the results are included in Section 5.

77 2. Datasets and methods

78 2.1 data descriptions

79 The global satellite-based dataset of monthly snow concentrations was provided by Rutgers University 80 (Robinson et al., 1993). Based on the daily product of the Interactive Multisensor Snow and Ice Mapping System, 81 monthly 89×89 grid cell arrays of snow data were generated. To examine the reliability of this reanalysis snow 82 data, routine daily snow observations at meteorological stations were also used (Bulygina et al., 2011) and were 83 downloaded from the website http://meteo.ru/tech/aisori.php. Considering the available timescale of the data, 421 84 stations were selected to collect data for the time period of 1980-2012 after quality control. Monthly sea ice (SI) 85 concentrations with a horizontal resolution of 1°×1° were downloaded from the Met Office Hadley Centre (Rayner et al., 2003) and these data are widely used in sea ice-related analysis. 86

The Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA2) is a NASA atmospheric reanalysis in the satellite era using the Goddard Earth Observing System Model, Version 5 (GEOS-5) with its Atmospheric Data Assimilation System (ADAS). The meteorological fields data with a horizontal 90 resolution of 0.5° latitude by 0.625° longitude were taken from the MERRA2 dataset (Gelaro et al., 2017), 91 including the geopotential height (Z) at 500 hPa and wind at 850 hPa, surface air temperature (SAT) and wind, 92 area fraction of middle and low clouds, boundary layer height (BLH), air temperature at 200 hPa, surface incoming 93 shortwave flux, surface net shortwave radiation, surface net longwave radiation, surface sensible heat flux, surface 94 latent heat flux and precipitation. These monthly mean MERRA2 data spanning from 1980 to 2018 were derived 95 from the Goddard Earth Sciences Data and Information Services Center. Besides, the abovementioned 96 atmospheric variables were also downloaded from the fifth generation European Center for Medium Range 97 Weather Forecasts (Copernicus Climate Change Service, 2017) to repeat the observational analyses and confirm 98 the robustness of the conclusions. Modified from Wang and He (2015), According to Yin et al. (2020a), the 99 calculatedion of the summer EU index-was as follows:

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$$EU = \left[-1 \times \overline{Z500}_{(59-75^{\circ}N, 66-100^{\circ}E)} + \overline{Z500}_{(40-54^{\circ}N, 105-128^{\circ}E)} - 1 \times \overline{Z500}_{(27-33^{\circ}N, 126-137^{\circ}E)} \right] / 3$$

101 where Z500 represents the geopotential height at 500 hPa and overbars denote the area average.

Ground-level O₃ concentrations have been observed since 2014 in China and are not sufficient to find longterm standing climate relationships. In this study, we employed the ozone weather index (OWI) during 1980– 2018, which has been defined by Yin et al. (2019; 2020b) and was proven to be a comprehensive and effective index determining the maximum daily average 8-h concentration of ozone (MDA8 O₃). The correlation coefficient between the observed MDA8 and daily OWI was 0.61 for the period 2007-2017 (Yin et al., 2019). The formula for OWI in North China is as follows:

108 OWI = normalized V10mI + normalized BI – normalized PI + normalized DTI.

109 where the V10mI is the area-averaged meridional wind at 10 m (35°-50°N, 110°-122.5°E), the BI is the area-110 averaged boundary layer height (37.5°-47.5°N, 112.5°-120°E), the PI is the area-averaged precipitation (37.5°-111 42.5°N, 112°-127.5°E), and the DTI is the area-averaged difference between the temperature at the surface 112 (37.5°-47.5°N, 110°-122.5°E) and at 200 hPa (37.5°-50°N, 110°-127.5°E). The normalized process is to divide 113 the anomaly by the standard deviation. These meteorological factors were selected based on their physicochemical 114 impacts on MDA8 O3 that were summarized in Figure S2. For example, (1) anomalous southerlies (expressed by 115 V10mI) transported O₃ precursors from Yangtze River Delta and superposed them with the local high emissions 116 in North China (Yin et al., 2019; Gong et al., 2020); (2) More precipitation indicated more cloud cover and stronger 117 efficiency of sunlight blocking-and wet removal (-PI); (3) Cooler high-level troposphere corresponded to 118 anticyclonic anomalies and sunny sky, and warmer surface air and higher BLH resulted in active natural emissions

119 of precursors and photochemical reaction (DTI, BI).

120 **2.2 GEOS-Chem simulations**

121 To verify the statistical physical mechanisms and fill the gap between OWI and MDA8 O₃, numerical 122 simulations based on the nested version of global 3-D chemical transport model (GEOS-Chem) were designed 123 and carried out. The GEOS-Chem model includes fully coupled O3-NOx-hydrocarbon and aerosol chemistry with 124 more than 80 species and 300 reactions (Bey et al., 2001), and is driven by the MERRA2 meteorological data with 125 $0.5^{\circ} \times 0.625^{\circ}$ horizontal resolution and 47 vertical levels over nested grid over Asia (11°S–55°N, 60°E–150°E). 126 The simulated ozone concentrations and the mass fluxes of ozone were calculated during the GEOS-Chem 127 simulations. Now there are six major components (i.e., chemical reaction, transport, PBL mixing, convection, 128 emissions and dry deposition, wet deposition) implemented for the budget diagnostics in GEOS-Chem model. 129 Because non-local planetary boundary layer (PBL) mixing was used in the simulation, the emissions and dry 130 deposition trends below the PBL were included within the mixing (Holtslag et al., 1993). Compared with other 131 terms, the value of wet deposition was extremely small, so it was not considered in this study (Liao et al., 2006). 132 Consequently, the major physical-chemical processes connected with meteorological conditions included the 133 chemical reaction, transport, PBL mixing, convection and their sum within the PBL.

134 In this study, the GEOS-Chem model was driven by changing meteorological conditions during 1980–2018 135 but with fixed anthropogenic emissions (MIX emission inventory in 2010) including from industry, power, residential and transportation sectors (Li et al., 2017); therefore, the interannual variations in MDA8 O₃ were 136 137 mainly caused by meteorological anomalies. The simulated MDA8 O₃ were analyzed in two ways depending on 138 two indexes (e.g., the years with the highest indexes minus those with the lowest indexes). The first composite 139 was designed to investigate the sustaining impacts of the EU pattern on MDA8 O_3 in North China (EX_{EU}) and the 140 differences of simulated results between six highest and six lowest EU index years were calculated during 1980-141 2018. The second composite attempted to verify the changing influences of April-May (AM) snow cover on 142 MDA8 O₃ (EX_{SC}). The EX_{SC} was executed in two separate periods: 1980–1998 and 1999–2018. In each sub-143 period, the simulated MDA8 O₃ was composited between the three lowest and three highest years of snow cover 144 anomaly values.

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3. Robust and <u>C</u>changing connection <u>between OWI and SCws</u>s

MDA8 O₃ highly correlated with the meteorological conditions. Yin et al. (2019) developed an index termed
 OWI to simulate the O₃ variations in North China (see Section 2.1) and largely extended the study period of O₃
 pollution. Although the calculations of OWI were constructed based on the datasets from 2006–2016 in a regional

149 background air-monitoring station (located at 40.7°N, 117.1°E; and 293.3ma.m.s.l), it is evident that OWI stably 150 reproduced the interannual variation in observed MDA8 O₃ in North China from 2014 to 2018 (green line in 151 Figure 1a). Thus, the summer-mean OWI can be used to indicate the joint effects of O₃-related meteorology in the 152 interannual time scale. Furthermore, GEOS-Chem model was driven by meteorological conditions from 1980 to 153 2018 with a fixed emission level. The simulated MDA8 O_3 showed similar interannual variations with the 154 observations during 2014–2018 after removal of the linear trend (Figure S3), indicating good performances of our 155 GEOS-Chem simulations. The MDA8 O₃ from GEOS-Chem mainly reflects the impacts of meteorological 156 variability on surface O_3 via modulating the dispersions, photochemical productions and meteorology-emission 157 interactions (Dang et al., 2020). The correlation coefficient between the observed JJA-mean OWI and simulated 158 MDA8 O₃ was 0.6 from 1980 to 2018 (above the 99% confidence level) and the 21-yr running correlation 159 coefficients maintained around 0.7 (Figure S4). The extreme OWI anomalies in 1990, 1997–1999, 2007, 2014 160 and 2017 were also consistent with the results of the GEOS-Chem simulations (Figure 1a). Therefore, the observed 161 OWI agreed with simulated MDA8 O₃ and successfully reflected the variation in O₃-related meteorology and its 162 impacts on O₃ pollution in North China.

163 As aforementioned, the positive phase of the EU pattern was found to have a close relationship with the interannual variations in the OWI (Yin et al., 2019); the correlation coefficient was 0.65 from 1980 to 2018 after 164 165 detrending (Figure 1a). In the 13 years when OWI reached extreme values (i.e., |OWI| > 1 × standard deviation), 166 the EU pattern also showed large values (i.e., $|EU| > 0.8 \times$ standard deviation) in 8 years, accounting for 62% of the larger OWI anomalies. The correlation coefficient between the EU index and simulated MDA8 O₃ (i.e., 0.56) 167 also exceeded the 99% confidence level during 1980–2018. In the $\mathrm{EX}_{\mathrm{EU}}$ experiment, the simulated MDA8 O_3 168 169 values in the six years with the highest and the six years with the lowest EU indexes were composited (highest 170 minus lowest). Because emissions fixed, the significantly positive anomalies of MDA8 O₃ in Figure 1c resulted 171 from different phases of the EU teleconnection and verified the impacts of the EU pattern on O₃ pollution in North 172 China. The physical-chemical processes of ozone production in GEOS-Chem simulations were analyzed. When 173 the EU pattern was at high positive phase, chemical reactions had large positive values. Although transport and 174 mixing had negative values, the sum of all physical-chemical processes was 8.27 Tons d⁻¹, resulting more O₃ 175 (Figure 1d). Furthermore, the 21-year running correlation coefficient between the EU index and observed OWI 176 (simulated MDA8 O₃) remained at approximately 0.7 (0.6) and was persistently above the 99% confidence level 177 (Figure 1b), indicating that the connections between the EU pattern and O₃-related meteorology in North China 178 did not change over time.



180 Figure 1. (a) The normalized variation in JJA-mean OWI (black), EU index (blue), simulated MDA8 O₃ (red) from 1980 to 181 2018 and observed MDA8 O₃ (green) from 2014 to 2018 after detrending. (b) The 21-year sliding correlation coefficients 182 between simulated MDA8 O₃ (red), OWI (black) and EU. The black dotted line (crosses) indicates (exceeded) the 95% 183 confidence level. (c) Composite difference of the simulated MDA8 O_3 (unit: $\mu g m^{-3}$) in summer between the six highest and 184 the six lowest EU index years from 1980 to 2018. The white dots (hatching) indicate that the difference was above the 95% 185 (90%) confidence level (t test). The green box represents the location of North China. (d) Composite difference of the mass 186 fluxes of summer ozone (unit: tons d⁻¹) from the GEOS-Chem between the six highest and the six lowest EU years from 1980 187 to 2018. The left axis is the name of the physical-chemical processes: chemical reaction (Chem), transport (Trans), PBL mixing 188 (Mix), convection (Conv) and their sums (Sum).

189 The 39-yr correlation coefficients between AM-mean Eurasia snow cover and summer mean OWI were 190 weakly negative (figure omitted). However, they were significantly negative in West Siberia and Central Siberia 191 during 1980–1998 (P1, Figure 2a) and these correlations disappeared during the period of 1999–2018 (P2, Figure 192 2b). The availability of snow data in three regions (i.e., West Siberia, Central Siberia and the northern area to 193 Baikal) was verified before confirming the key region of snow cover anomalies. Judging from the spatial and 194 temporal correlation analysis, the reanalysis data of snow cover provided by Rutgers University agreed well with 195 the site observations in West Siberia (62°-66°N, 75°-92°E) from 1980 to 2012 (Figure S5). Thus, the regional 196 mean of AM-mean Eurasian snow cover in this region was defined as the SC_{WS}, which was also significantly and 197 negatively correlated with the summer EU pattern (Figure S6). Furthermore, as pointed by Yin et al. (2020a), sea 198 ice anomalies in the Gakkel Ridge (SI_{GR}, 82°-88°N, 0°-80°E, Figure S1) also bridged the summer EU and OWI.



Figure 2. The correlation coefficients between the JJA-mean OWI and AM-mean snow cover (a) from 1980 to 1998 and (b) from 1999 to 2018. The white dots (hatching) indicate that the correlation coefficients exceeded the 95% (90%) confidence level (*t* test). The green box represents the key area used to calculate the SCsw index. The linear trend is removed.

203 During 1980–2018, the correlation coefficients between OWI and the above two external forcings were 0.5 (SI_{GR}, significant at the 99% confidence level) and -0.21 (SC_{WS}, insignificant at the 95% confidence level), 204 205 respectively (Figure 3 a, b). We also checked the 21-year running correlation coefficients between each forcing 206 and OWI in Figure 3c-d, both of which showed decadal changes and independent with the choice of running time 207 window (Figure omitted). The correlation between OWI and SC_{WS} was significant (-0.68, above the 99% 208 confidence level) during P1 and became insignificant (0.20) during P2 (Figure 3c). Oppositely, the correlation 209 with SIGR enhanced from 0.4 in P1 to 0.62 in P2 (Figure 3d). After removing the signal of El Niño-Southern 210 Oscillation (ENSO), these correlation coefficients almost unchanged. Interestingly, the connections between these 211 two preceding factors and the EU pattern illustrated similar decadal changes (Figure 3 c, d). That is, the correlation 212 between EU and SC_{WS} was only significant (-0.62) in the former period; however, the correlation between EU 213 and SIGR was only significant (0.61) after the mid-1990s (Figure 3 c, d). Furthermore, the SIGR and SCWS were 214 mutually independent because the 21-year running correlation coefficient between them was maintained at a low 215 level (Figure S7). Therefore, we speculated that the impacts of the summer EU pattern on ground-level O_3 216 pollution in North China were robust and long-standing (Figure 1b). However, the preceding factors inducing the 217 EU pattern to influence the O3 pollution in North China changed from SC_{WS} in P1 to SI_{GR} in P2 (Figure 3 c, d).



Figure 3. The normalized variation in (a) OWI (black) and SCws (red), (b) OWI (black) and SI_{GR} (blue) from 1980 to 2018 after detrending. The 21-year sliding correlation coefficients between (c) SCws and OWI (red), EU (blue), (d) SI_{GR} and OWI (red), EU (blue). The black dotted line (crosses) indicates (exceeded) the 95% confidence level. The linear trend is removed.

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4. Possible Pphysical mechanisms

223 The physical mechanisms of how to achieve the impacts of SC_{WS} on surface O_3 pollution in North China is 224 still a new question to the best of our knowledge. As an efficient climate forcing, the snow cover anomalies could 225 stimulate synchronous responses in the atmosphere by changing albedo and hydrological effects and could then 226 impact the atmosphere in the following seasons (Cohen and Rind, 1991). In April and May, the snow at high 227 latitudes began to melt and had obvious interannual variations, as shown by both the observations and the 228 reanalysis data (Figure S5). Generally, lower albedo, associated with less snow cover, meant that the land/snow 229 surface reflected less solar radiation and resulted in higher SAT. Warmer surfaces produce stronger longwave 230 radiation and heat the local atmosphere from the surface to the mid-troposphere (Chen et al., 2003; Chen et al., 231 2016). Moreover, the changing local soil moisture enhanced the surface heat flux and thus resulted in higher SAT and atmospheric temperatures (Zhang et al., 2017). Finally, the warmed thermal conditions in the atmosphere 232 233 enhanced the local 1000-500 hPa thickness and represented positive anomalies of Z500 (Chen et al., 2003; Halder 234 and Dirmeyer, 2017). Compared to P1, the south edge of the area with high concentrations of snow (>85%) in late 235 spring shifted northward by approximately 2° in latitude during P2 (Figure 4a). Similarly, the significant changes 236 in radiation flux (shortwave + longwave) and heat flux (latent + sensible) also moved northward in P2 relative to P1 (Figure 4 b, c). We speculated that this northward movement of effective snow cover, accompanied by shifts 237 238 in net heat flux, possibly contributed to the changing relationship between the SC_{WS} and OWI.



Figure 4. (a) The south edge of the 85% snow cover concontration during 1980–1993 (black) and during 2004–2018 (red). The gray (green) box represents the key area used to calculate the NHFws (SCsw) index. The correlation coefficients between the SCws× -1 and (b) surface net radiation flux (shortwave+longwave) and (c) surface net heat flux (latent+sensible) are displayed during 1980–1998 (shading) and 1999–2018 (contour). White dots (hatching) indicate that the correlation coefficients during P1 exceeding the 95% (90%) confidence level (*t* test). The gray (black) contours represent the correlation coefficients during P2 exceeding the 95% (90%) confidence level. The linear trend is removed.

246 The local responses of geopotential height in the mid-troposphere induced by negative anomalies of the SC_{WS} illustrated decadal changes; that is, the significant correlation coefficients between SC_{WS} \times -1 and Z500, as well 247 248 as SAT, were distributed more southward and were stronger in P1 (Figure 5a) than in P2 (Figure 5c). For 249 convenience, the roles of radiation and heat flux (shortwave + longwave + latent + sensible) were considered 250 together as net heat flux (Zhang et al., 2017), which was averaged over West Siberia (54°-68°N, 75°-92°E) and 251 defined as NHF_{WS}. It was evident that the atmospheric responses associated with the NHF_{WS} agreed well with 252 those of less SC_{WS} (Figure 5 b, d). That is, the enhanced net heat flux related to decreased snow cover in West 253 Siberia heated the above atmosphere and resulted in local warmer SAT and anticyclonic circulations in the mid-254 troposphere during P1 (Figure 5 a, b). In addition, cyclonic responses can be found on the left and right sides of 255 the aforementioned anticyclonic anomalies in April-May (Figure 5 a, b). However, similar to the radiation and 256 heat flux in Figure 4 b-c, the atmospheric responses were distributed more northward and were weaker during P2 257 than during P1 (Figure 5 c, d).



Figure 5. The correlation coefficients between the $SC_{WS} \times -1$ (a, c), NHF_{WS} (b, d) and surface air temperature (shading) and geopotential height at 500 hPa (contour) from 1980 to 1998 (a, b) and from 1999 to 2018 (c, d). The white dots (hatching) indicate that the correlation coefficients in shading exceeded the 95% (90%) confidence level (*t* test). The green boxes represent the anomalous cyclonic or anticyclonic centers in AM. The linear trend is removed.

263 The AM-mean NHF_{WS} showed significantly positive correlations with both the summer-mean EU (0.49) and OWI (0.52) during P1 (Figure 6a, S8a). After removing the ENSO signal, these correlated relationships almost 264 265 showed no difference. Furthermore, the "-+-" anomalous atmospheric centers in April-May (green boxes in 266 Figure 5a) had significantly positive correlations with the summer EU pattern (CC=0.45, above the 95% confidence level). The atmospheric anomalies stimulated by negative SC_{WS} could appear as positive phases of the 267 268 EU pattern in JJA during P1 (Figure 6b, S8a). As one center of the EU pattern, the anticyclonic anomalies over 269 North China were significant in the mid- and lower-troposphere (Figure 6b, 7a) and resulted in clear skies (Figure 270 7c). Sinking heating, intense sunlight (Figure 7c) and less precipitation (correspondingly more cloud and weaker ultraviolet radiations, Figure 7a) resulted in beneficial environments for the natural emissions of O₃ precursors 271 272 (Lu et al., 2019) and photochemical reactions (Pu et al., 2017). Differently, the northward and weaker atmospheric 273 responses in April-May were almost dispersed in summer (Figure 6c, S8b) and had little impacts on the local OWI 274 in North China (Figure 7 b, d) during P2, which were consistent with the insignificant correlations between the 275 NHF_{ws} and the EU (OWI) (Figure 6a).



Figure 6. (a) The normalized variation in the JJA OWI (black), JJA EU index (red) and AM NHF_{ws} (blue) from 1980 to 2018 after detrending. The numbers represent the correlation coefficients between the NHF_{ws} and EU, OWI during 1980–1998 and 1999–2018, respectively. Two asterisks indicate that the correlation coefficients exceeded the 95% confidence level. The correlation coefficients between $SC_{ws} \times -1$ and JJA surface air temperature (shading) and geopotential height at 500 hPa (contour) from 1980 to 1998 (b) and from 1999 to 2018 (c). The white dots (hatching) indicate that the correlation coefficients with surface air temperature exceeded the 95% (90%) confidence level (*t* test). The green boxes represent the key areas used to calculate the EU index. The linear trend is removed.





Figure 7. The meteorological conditions associated with the $SC_{WS} \times -1$. (a, b) The correlation coefficients between $SC_{WS} \times -1$ and precipitation (shading) and wind at 850 hPa (arrow); (c, d) surface incoming shortwave flux (shading), and the sum of low and medium cloud cover (contour) from 1980 to 1998 (a, c) and from 1999 to 2018 (b, d). The white dots (hatching) indicate

that the correlation coefficients represented with shading exceeded the 95% (90%) confidence level (*t* test). The gray (black)
contours exceeded the 95% (90%) confidence level. The green boxes represent the location of North China. The linear trend
is removed.

291 In the EX_{SC} experiment, the simulated MDA8 O₃ and mass fluxes of ozone were composited (three lowest SC_{WS} minus highest) during P1 and P2, respectively. During P1, the composited results (with fixed emissions) 292 293 were significantly positive (Figure 8a) and were in good agreement with the proposed mechanisms (i.e., less snow 294 cover in West Siberia resulted in severe surface O₃ pollution in North China). The responses of MDA8 O₃ pollution 295 in North China were insignificant during P2 (Figure 8b) and were also consistent with both weak impacts in this 296 period and changing relationships. Mass balance of ozone are jointly determined by four processes (i.e., chemistry, 297 transport, PBL mixing and convection) which could be isolated by the GEOS-Chem model. During P1, the 298 composite results of chemical reaction had large positive values (11.05 Tons d^{-1}) (Figure 8a), indicating that the 299 dry-hot meteorological conditions were conductive to produce more O₃. Anomalous anticyclonic circulations 300 located above the North China region resulted in downward air flow that may bring the ozone from the 301 stratosphere to surface. Hence, the value of convection was also positive. The values of transport and mixing were 302 negative (Figure 8a), but the sum of all processes was positive, indicating the ozone concentrations in North China 303 would increase. However, the composite results of chemical, transport and mixing were opposite (Figure 8b) 304 during P2 compared with P1. Meanwhile, the values of convection and the sum were extremely close to zero 305 (Figure 8b), indicating that there were little impacts on ozone in North China when the SC_{WS} was low during P2. 306 The composite results of mass fluxes were well agreement with the previous conclusion.





315 In this study, the April-May snow cover in West Siberia was newly proposed as a preceding climate driver 316 that influenced the surface O₃-related meteorology in North China during 1980–1998, and the associated physical mechanisms were also explained by comparing the periods before and after the mid-1990s. Accompanying the 317 northward shift of dense snow cover, the associated radiation and heat flux also retreated toward the polar region 318 319 during 1999–2018 (Figure 4); thus, the induced atmospheric anomalies were located northward in April-May and 320 disappeared in summer (Figure 5 c, d). However, in the period of 1980–1998, the positive phase of the EU pattern 321 in summer could be stimulated by negative anomalies of snow cover (mainly by enhanced net heat flux) in West 322 Siberia (Figure 6). Consequently, hot-dry air and intense solar radiation under anomalous anticyclonic circulations 323 not only enhanced the natural emissions of O₃ precursors but also promoted photochemical reactions to produce 324 more O₃ near the surface (Figure 7, 8). To enhance the robustness of this study, the ERA5 reanalysis data were 325 also employed to reproduce the observational analyses. As shown in Figure S9, identical results were obtained 326 and confirmed.

327 The linkage between the EU pattern and MDA8 O_3 was robust, which bridged the SC_{WS} and OWI in the 328 period of 1980–1998 but connected the SI_{GR} and OWI after the mid-1990s. In Figure 9, the OWI were regressed 329 by SC_{WS} and SI_{GR} from 1980–2018. The 21-year running correlation coefficient between the OWI and the fitted 330 values stably maintained around 0.6 and indicated that these two preceding factors almost introduced the full impacts of the EU pattern (Figure 1b) over the whole period. Generally, the decadal changes in the climate drivers 331 332 influences the stability of the predictability. It is evident that our results overcame this problem and deepened the 333 understanding of variations in summer O₃ from the climate perspective. Yin et al. (2020a) also found that the sea ice anomalies over the Canada Basin and the Beaufort Sea (Figure S1) also stimulated a Rossby-wave-like train 334 335 propagating through the North Pacific to influence the variability in the OWI in North China. When we added 336 these sea ice anomalies into the regressions, the fitting performance was visibly improved because the 21-year 337 running correlation coefficient was elevated to approximately 0.8 with OWI, as seen in Figure 9b.



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Figure 9. (a) The variation in the JJA-mean observed OWI (black), the fitted OWI-1 (by the SC_{WS} and SI_{GR}, red), and the fitted OWI-2 (by the SC_{WS}, SI_{GR} and SI_{CB}, blue) from 1980 to 2018 after detrending. (b) The 21-year sliding correlation coefficients between observed OWI and fitted OWI-1 (red), fitted OWI-2 (blue). The black dotted line (crosses) indicates (exceeded) the 95% confidence level.

343 The concentrations of surface O₃ have been extensively measured since 2014 in China; this time scale cannot support the study of the interannual-decadal variability in O_3 pollution. In this study, we used two datasets, i.e., 344 345 the ozone weather index and the O₃ concentrations simulated by GEOS-Chem, to focus on the impacts of climate 346 variability on surface O_3 in North China. Although the feasibility of these datasets was strictly examined, there 347 were still gaps between the real variations in O_3 and the variations in these two substitutions; this discrepancy 348 requires further research. Moreover, the climate-chemistry coupled model need be used to verify the role of snow 349 cover on ozone pollution in North China in further studies. Furthermore, there is no doubt that anthropogenic 350 emissions are the fundamental drivers of O₃ pollution, which has been investigated in many previous studies (Li 351 et al., 2018; Li et al., 2019; Dang et al., 2020). After removal of the linear trend, the signals of climate warming 352 in the atmosphere were also eliminated, which allowed us to focus on the interannual variations. In addition, the decrease in haze aerosols was also proven to be an effective contributor to recent interannual variations in O₃ 353 354 concentrations (Li et al., 2019), which were not involved in our study and need further attentions.

355 Data Availability. Hourly O₃ concentration data can be downloaded from https://quotsoft.net/air/ (Ministry of 356 Environmental Protection of China, last accessed on 8 November 2020). Sea ice concentration data are from 357 https://www.metoffice.gov.uk/hadobs/hadisst/data/download.html (Met Office Hadley Centre, 2020). Snow cover 358 data can be downloaded from Rutgers University at http://climate.rutgers.edu/snowcover/ (Rutgers University, 359 2020). The observed snow data from meteorological stations are available at http://meteo.ru/tech/aisori.php. The 360 monthly mean MERRA2 reanalysis datasets are available at https://disc.gsfc.nasa.gov/datasets?page=1 (last 361 access: 21 March 2021). The monthly mean ERA5 reanalysis datasets are available at https://cds.climate.copernicus.eu/cdsapp#!/home (Copernicus Climate Change Service, last accessed on 9 362 363 November 2020).

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367 Authors' contribution

Wang H. J. and Yin Z. C. designed and performed researches. Wan Y. did the statistical analysis and implemented
the GEOS-Chem simulations. Yin Z. C. prepared the manuscript with contributions from all co-authors.

370 Competing interests

371 The authors declare no conflict of interest.

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479 Table and Figure captions

Figure 1. (a) The normalized variation in JJA-mean OWI (black), EU index (blue), simulated MDA8 O₃ (red) 480 from 1980 to 2018 and observed MDA8 O₃ (green) from 2014 to 2018 after detrending. (b) The 21-year sliding 481 correlation coefficients between simulated MDA8 O₃ (red), OWI (black) and EU. The black dotted line (crosses) 482 483 indicates (exceeded) the 95% confidence level. (c) Composite difference of the simulated MDA8 O_3 (unit: $\mu g m^{-3}$) 484 in summer between the six highest and the six lowest EU index years from 1980 to 2018. The white dots (hatching) 485 indicate that the difference was above the 95% (90%) confidence level (t test). The green box represents the 486 location of North China. (d) Composite difference of the mass fluxes of summer ozone (unit: tons d⁻¹) from the 487 GEOS-Chem between the six highest and the six lowest EU years from 1980 to 2018. The left axis is the name of 488 the physical-chemical processes: chemical reaction (Chem), transport (Trans), PBL mixing (Mix), convection 489 (Conv) and their sums (Sum).

- 490 Figure 2. The correlation coefficients between the JJA-mean OWI and AM-mean snow cover (a) from 1980 to 491 1998 and (b) from 1999 to 2018. The white dots (hatching) indicate that the correlation coefficients exceeded the 492 95% (90%) confidence level (t test). The green box represents the key area used to calculate the SCsw index. The 493 linear trend is removed.
- Figure 3. The normalized variation in (a) OWI (black) and SC_{WS} (red), (b) OWI (black) and SI_{GR} (blue) from
 1980 to 2018 after detrending. The 21-year sliding correlation coefficients between (c) SC_{WS} and OWI (red), EU
 (blue), (d) SI_{GR} and OWI (red), EU (blue). The black dotted line (crosses) indicates (exceeded) the 95%
 confidence level. The linear trend is removed.
- **Figure 4.** (a) The south edge of the 85% snow cover concontration during 1980–1993 (black) and during 2004– 2018 (red). The gray (green) box represents the key area used to calculate the NHF_{ws} (SCsw) index. The correlation coefficients between the SC_{ws}× -1 and (b) surface net radiation flux (shortwave+longwave) and (c) surface net heat flux (latent+sensible) are displayed during 1980–1998 (shading) and 1999–2018 (contour). White dots (hatching) indicate that the correlation coefficients during P1 exceeding the 95% (90%) confidence level (t test). The gray (black) contours represent the correlation coefficients during P2 exceeding the 95% (90%) confidence level. The linear trend is removed.
- 505 Figure 5. The correlation coefficients between the $SC_{WS} \times -1$ (a, c), NHF_{WS} (b, d) and surface air temperature
- 506 (shading) and geopotential height at 500 hPa (contour) from 1980 to 1998 (a, b) and from 1999 to 2018 (c, d).
- 507 The white dots (hatching) indicate that the correlation coefficients in shading exceeded the 95% (90%) confidence 508 level (t test). The green boxes represent the anomalous cyclonic or anticyclonic centers in AM. The linear trend is
- 509 removed.
- 510 Figure 6. (a) The normalized variation in the JJA OWI (black), JJA EU index (red) and AM NHF_{ws} (blue) from
- 511 1980 to 2018 after detrending. The numbers represent the correlation coefficients between the NHF_{WS} and EU,
- 512 OWI during 1980–1998 and 1999–2018, respectively. Two asterisks indicate that the correlation coefficients

- 513 exceeded the 95% confidence level. The correlation coefficients between $SC_{WS} \times -1$ and JJA surface air
- 514 temperature (shading) and geopotential height at 500 hPa (contour) from 1980 to 1998 (b) and from 1999 to 2018
- 515 (c). The white dots (hatching) indicate that the correlation coefficients with surface air temperature exceeded the
- 516 95% (90%) confidence level (t test). The green boxes represent the key areas used to calculate the EU index. The
- 517 linear trend is removed.
- 518 Figure 7. The meteorological conditions associated with the $SC_{WS} \times -1$. (a, b) The correlation coefficients
- 519 between $SC_{WS} \times -1$ and precipitation (shading) and wind at 850 hPa (arrow); (c, d) surface incoming shortwave
- 520 flux (shading), and the sum of low and medium cloud cover (contour) from 1980 to 1998 (a, c) and from 1999 to
- 521 2018 (b, d). The white dots (hatching) indicate that the correlation coefficients represented with shading exceeded
- 522 the 95% (90%) confidence level (t test). The gray (black) contours exceeded the 95% (90%) confidence level. The
- 523 green boxes represent the location of North China. The linear trend is removed.
- 524 Figure 8. Composite difference of the summer MDA8 O₃ (unit: µg m-3) simulated by the GEOS-Chem model
- between the three lowest and the three highest SC_{WS} years (a) from 1980 to 1998 and (b) from 1999 to 2018. The
- 526 white dots (hatching) indicate that the difference was above the 95% (90%) confidence level (t test). The green
- boxes represent the location of North China. The bar chart on the right is the composite difference of the summer
- 528 mass fluxes of ozone (unit: tons d-1) during each periods. The left axis is the name of the physical-chemical
- 529 processes: chemical reaction (Chem), transport (Trans), PBL mixing (Mix), convection (Conv) and their sums
- 530 (Sum). The results were calculated within the planetary boundary layer.
- 531 Figure 9. (a) The variation in the JJA-mean observed OWI (black), the fitted OWI-1 (by the SC_{WS} and SI_{GR}, red),
- and the fitted OWI-2 (by the SC_{WS} , SI_{GR} and SI_{CB} , blue) from 1980 to 2018 after detrending. (b) The 21-year
- 533 sliding correlation coefficients between observed OWI and fitted OWI-1 (red), fitted OWI-2 (blue). The black
- 534 dotted line (crosses) indicates (exceeded) the 95% confidence level.