



1	Decadal Changes of Connections among Snow cover in West Siberia, Eurasia
2	Teleconnection and O <sub>3</sub> -related meteorology in North China
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18	Abstract. Severe surface ozone $(O_3)$ pollution frequently occurred in North China and obviously damages human
19	health and ecosystems. The meteorological conditions effectively modulate the variations in $\mathrm{O}_3$ pollution. In this
20	study, the interannual relationship between $O_3$ -related meteorology and late-spring snow cover in West Siberia
21	was explored, and the reasons of its decadal change were also physically explained. Before mid-1990s, less snow
22	cover could enhance net heat flux and stimulate positive phase of the Eurasia (EU) teleconnection in summer. The
23	positive EU pattern resulted in hot-dry air and intense solar radiation in North China, which could enhance the
24	natural emissions of $\mathrm{O}_3$ precursors and photochemical reactions in the atmosphere closely related to high $\mathrm{O}_3$
25	concentrations. However, after the mid-1990s, the south edge of the dense snow cover area in West Siberia shifted
26	northward by approximately $2^{\circ}$ in latitude and accompanied radiation and heat flux also retreated toward the polar
27	region. The connections among snow anomalies, EU pattern and surface $\mathrm{O}_3$ became insignificant and thus
28	influenced the stability of the predictability.

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





## 30 1. Introduction

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

49 High concentrations of ground-level ozone (O<sub>3</sub>) are frequently observed together with dry-hot air and intense 50 solar radiation because photochemical reactions are accelerated under such meteorological conditions (Pu et al., 51 2017). The large-scale atmospheric circulations associated with high-O<sub>3</sub>-related meteorology in North China 52 appeared as the positive phase of the EU pattern (Yin et al., 2019, 2020a). The anomalous anticyclonic circulations 53 over North China, as one active center of the EU pattern, induced significant descending air flows and thus 54 efficient adiabatic heating and intense sunlight (Gong and Liao, 2019). Generally, numerous nitrogen oxides (NOx) 55 and volatile organic compounds (VOCs) are emitted by human activities and natural sources in North China 56 (Zheng et al., 2018). These precursors of O<sub>3</sub> react under high ultraviolet radiation and generate more O<sub>3</sub> (Fix et 57 al., 2018).

58 The variation in the EU pattern and its linkage with surface O<sub>3</sub> in North China were both driven by preceding 59 spring forcings (Zhang et al., 2018; Yin et al., 2019, 2020a). Arctic sea ice anomalies in spring were proven to be





60 closely related to the summer EU teleconnection pattern; these anomalies then influenced rainfall in China (Wu 61 et al., 2009). Summer surface  $O_3$  in North China closely linked to the variability in May sea ice over the Gakkel 62 Ridge (Figure S1) and the bridge in atmosphere was the EU pattern (Yin et al., 2019, 2020a). However, this 63 relationship between sea ice anomalies and EU pattern showed a decadal change from insignificant to significant 64 after the mid-1990s (Yin et al., 2020a). The east-west dipole of spring snow cover anomalies in Eurasia was 65 closely related to the East Asia summer monsoon by stimulating atmospheric responses such as the EU pattern 66 (Yim et al., 2010). When building a seasonal prediction model of surface O<sub>3</sub>-related meteorology, the May snow 67 cover in West Siberia was selected as a predictor and effectively increased the predictability (Yin et al., 2020b). 68 However, the physical mechanisms linking O<sub>3</sub> and snow cover are still unclear.

Two open questions are as follows: (1) Have the links between the EU pattern and O<sub>3</sub>-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.

# 76 2. Datasets and methods

## 77 2.1 data descriptions

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

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





89 resolution of 0.5° latitude by 0.625° longitude were taken from the MERRA2 dataset (Gelaro et al., 2017), 90 including the geopotential height (Z) at 500 hPa and wind at 850 hPa, surface air temperature (SAT) and wind, 91 area fraction of middle and low clouds, boundary layer height (BLH), air temperature at 200 hPa, surface incoming 92 shortwave flux, surface net shortwave radiation, surface net longwave radiation, surface sensible heat flux, surface 93 latent heat flux and precipitation. These monthly mean MERRA2 data spanning from 1980 to 2018 were derived 94 from the Goddard Earth Sciences Data and Information Services Center. Besides, the abovementioned 95 atmospheric variables were also downloaded from the fifth generation European Center for Medium Range 96 Weather Forecasts (Copernicus Climate Change Service, 2017) to repeat the observational analyses and confirm 97 the robustness of the conclusions. According to Yin et al. (2020a), the calculation of the EU index was as follows:

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

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

Ground-level O<sub>3</sub> 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 O<sub>3</sub>). The formula for OWI in North China is as follows:

# 105 OWI = normalized V10mI + normalized BI – normalized PI + normalized DTI.

106 where the V10mI is the area-averaged meridional wind at 10 m  $(35^{\circ}-50^{\circ}N, 110^{\circ}-122.5^{\circ}E)$ , the BI is the area-107 averaged boundary layer height (37.5°-47.5°N, 112.5°-120°E), the PI is the area-averaged precipitation (37.5°-108 42.5°N, 112°-127.5°E), and the DTI is the area-averaged difference between the temperature at the surface 109 (37.5°-47.5°N, 110°-122.5°E) and at 200 hPa (37.5°-50°N, 110°-127.5°E). These meteorological factors were 110 selected based on their physicochemical impacts on MDA8 O<sub>3</sub> that were summarized in Figure S2. For example, 111 (1) anomalous southerlies (expressed by V10mI) transported  $O_3$  precursors from Yangtze River Delta and 112 superposed them with the local high emissions in North China; (2) More precipitation indicated stronger efficiency 113 of sunlight blocking and wet removal (-PI); (3) Cooler high-level troposphere corresponded to anticyclonic 114 anomalies and sunny sky, and warmer surface air and higher BLH resulted in active natural emissions of 115 precursors and photochemical reaction (DTI, BI).

#### 116 2.2 GEOS-Chem simulations

117 To verify the statistical physical mechanisms and fill the gap between OWI and MDA8 O<sub>3</sub>, numerical





118 simulations based on the nested version of global 3-D chemical transport model (GEOS-Chem) were designed 119 and carried out. The GEOS-Chem model includes fully coupled O3-NOx-hydrocarbon and aerosol chemistry with 120 more than 80 species and 300 reactions (Bey et al., 2001), and is driven by the MERRA2 meteorological data with 121 0.5°×0.625° horizontal resolution and 47 vertical levels over nested grid over Asia (11°S-55°N, 60°E-150°E). 122 The simulated ozone concentrations and the mass fluxes of ozone were calculated during the GEOS-Chem 123 simulations. Now there are six major components (i.e., chemical reaction, transport, PBL mixing, convection, 124 emissions and dry deposition, wet deposition) implemented for the budget diagnostics in GEOS-Chem model. 125 Because non-local planetary boundary layer (PBL) mixing was used in the simulation, the emissions and dry 126 deposition trends below the PBL were included within the mixing (Holtslag et al., 1993). Compared with other 127 terms, the value of wet deposition was extremely small, so it was not considered in this study (Liao et al., 2006). 128 Consequently, the major physical-chemical processes connected with meteorological conditions included the 129 chemical reaction, transport, PBL mixing, convection and their sum within the PBL.

130 In this study, the GEOS-Chem model was driven by changing meteorological conditions during 1980-2018 131 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 132 133 mainly caused by meteorological anomalies. The simulated MDA8 O<sub>3</sub> were analyzed in two ways depending on 134 two indexes (e.g., the years with the highest indexes minus those with the lowest indexes). The first composite 135 was designed to investigate the sustaining impacts of the EU pattern on MDA8 O<sub>3</sub> in North China (EX<sub>EU</sub>) and the 136 differences of simulated results between six highest and six lowest EU index years were calculated during 1980-137 2018. The second composite attempted to verify the changing influences of April-May (AM) snow cover on 138 MDA8 O<sub>3</sub> (EX<sub>SC</sub>). The EX<sub>SC</sub> was executed in two separate periods: 1980–1998 and 1999–2018. In each sub-139 period, the simulated MDA8 O<sub>3</sub> was composited between the three lowest and three highest years of snow cover 140 anomaly values.

#### 141 3. Robust and changing connections

MDA8 O<sub>3</sub> highly correlated with the meteorological conditions. Yin et al. (2019) developed an index termed OWI to simulate the O<sub>3</sub> variations in North China (see Section 2.1) and largely extended the study period of O<sub>3</sub> pollution. Although the calculations of OWI were constructed based on the datasets from 2006–2016 in a regional background air-monitoring station (located at 40.7°N, 117.1°E; and 293.3ma.m.s.l), it is evident that OWI stably reproduced the interannual variation in observed MDA8 O<sub>3</sub> in North China from 2014 to 2018 (green line in Figure 1a). Thus, the summer-mean OWI can be used to indicate the joint effects of O<sub>3</sub>-related meteorology in the



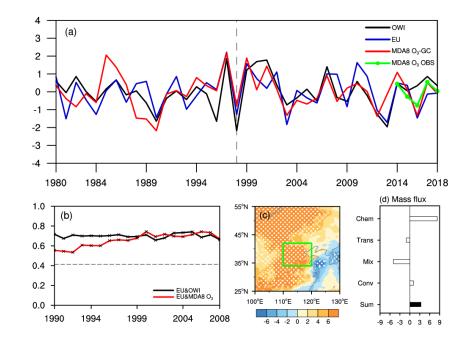


148 interannual time scale. Furthermore, GEOS-Chem model was driven by meteorological conditions from 1980 to 149 2018 with a fixed emission level. The simulated MDA8 O<sub>3</sub> showed similar interannual variations with the 150 observations during 2014-2018 after removal of the linear trend (Figure S3), indicating good performances of our 151 GEOS-Chem simulations. The MDA8 O3 from GEOS-Chem mainly reflects the impacts of meteorological 152 variability on surface O<sub>3</sub> via modulating the dispersions, photochemical productions and meteorology-emission 153 interactions (Dang et al., 2020). The correlation coefficient between the observed JJA-mean OWI and simulated 154 MDA8 O3 was 0.6 from 1980 to 2018 (above the 99% confidence level) and the 21-yr running correlation 155 coefficients maintained around 0.7 (Figure S4). The extreme OWI anomalies in 1990, 1997-1999, 2007, 2014 156 and 2017 were also consistent with the results of the GEOS-Chem simulations (Figure 1a). Therefore, the observed 157 OWI agreed with simulated MDA8 O<sub>3</sub> and successfully reflected the variation in O<sub>3</sub>-related meteorology and its 158 impacts on O<sub>3</sub> pollution in North China.

159 As aforementioned, the positive phase of the EU pattern was found to have a close relationship with the 160 interannual variations in the OWI (Yin et al., 2019); the correlation coefficient was 0.65 from 1980 to 2018 after 161 detrending (Figure 1a). In the 13 years when OWI reached extreme values (i.e., |OWI| > 1 × standard deviation), 162 the EU pattern also showed large values (i.e.,  $|EU| > 0.8 \times$  standard deviation) in 8 years, accounting for 62% of 163 the larger OWI anomalies. The correlation coefficient between the EU index and simulated MDA8 O<sub>3</sub> (i.e., 0.56) 164 also exceeded the 99% confidence level during 1980–2018. In the  $EX_{EU}$  experiment, the simulated MDA8  $O_3$ 165 values in the six years with the highest and the six years with the lowest EU indexes were composited (highest 166 minus lowest). Because emissions fixed, the significantly positive anomalies of MDA8 O<sub>3</sub> in Figure 1c resulted 167 from different phases of the EU teleconnection and verified the impacts of the EU pattern on O<sub>3</sub> pollution in North 168 China. The physical-chemical processes of ozone production in GEOS-Chem simulations were analyzed. When 169 the EU pattern was at high positive phase, chemical reactions had large positive values. Although transport and 170 mixing had negative values, the sum of all physical-chemical processes was 8.27 Tons  $d^{-1}$ , resulting more O<sub>3</sub> 171 (Figure 1d). Furthermore, the 21-year running correlation coefficient between the EU index and observed OWI 172 (simulated MDA8 O<sub>3</sub>) remained at approximately 0.7 (0.6) and was persistently above the 99% confidence level 173 (Figure 1b), indicating that the connections between the EU pattern and O<sub>3</sub>-related meteorology in North China 174 did not change over time.







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176 Figure 1. (a) The normalized variation in JJA-mean OWI (black), EU index (blue), simulated MDA8 O3 (red) from 1980 to 177 2018 and observed MDA8 O<sub>3</sub> (green) from 2014 to 2018 after detrending. (b) The 21-year sliding correlation coefficients 178 between simulated MDA8 O3 (red), OWI (black) and EU. The black dotted line (crosses) indicates (exceeded) the 95% 179 confidence level. (c) Composite difference of the simulated MDA8 O<sub>3</sub> (unit: µg m<sup>-3</sup>) in summer between the six highest and 180 the six lowest EU index years from 1980 to 2018. The white dots (hatching) indicate that the difference was above the 95% 181 (90%) confidence level (t test). The green box represents the location of North China. (d) Composite difference of the mass 182 fluxes of summer ozone (unit: tons d<sup>-1</sup>) from the GEOS-Chem between the six highest and the six lowest EU years from 1980 183 to 2018. The left axis is the name of the physical-chemical processes: chemical reaction (Chem), transport (Trans), PBL mixing 184 (Mix), convection (Conv) and their sums (Sum).

185 The 39-yr correlation coefficients between AM-mean Eurasia snow cover and summer mean OWI were 186 weakly negative (figure omitted). However, they were significantly negative in West Siberia and Central Siberia 187 during 1980-1998 (P1, Figure 2a) and these correlations disappeared during the period of 1999-2018 (P2, Figure 188 2b). The availability of snow data in three regions (i.e., West Siberia, Central Siberia and the northern area to 189 Baikal) was verified before confirming the key region of snow cover anomalies. Judging from the spatial and 190 temporal correlation analysis, the reanalysis data of snow cover provided by Rutgers University agreed well with 191 the site observations in West Siberia (62°-66°N, 75°-92°E) from 1980 to 2012 (Figure S5). Thus, the regional 192 mean of AM-mean Eurasian snow cover in this region was defined as the SC<sub>WS</sub>, which was also significantly and 193 negatively correlated with the summer EU pattern (Figure S6). Furthermore, as pointed by Yin et al. (2020a), sea 194 ice anomalies in the Gakkel Ridge (SI<sub>GR</sub>, 82°-88°N, 0°-80°E, Figure S1) also bridged the summer EU and OWI.





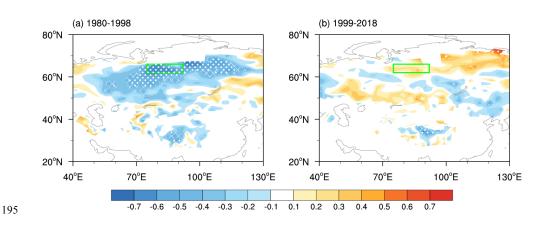


Figure 2. The correlation coefficients between the JJA-mean OWI and AM-mean snow cover (a) from 1980 to 1998 and (b) from 1999 to 2018. The white dots (hatching) indicate that the correlation coefficients exceeded the 95% (90%) confidence level (*t* test). The green box represents the key area used to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

199 During 1980-2018, the correlation coefficients between OWI and the above two external forcings were 0.5 200  $(SI_{GR}, significant at the 99\%$  confidence level) and -0.21 (SC<sub>WS</sub>, insignificant at the 95% confidence level), 201 respectively (Figure 3 a, b). We also checked the 21-year running correlation coefficients between each forcing 202 and OWI in Figure 3c-d, both of which showed decadal changes and independent with the choice of running time 203 window (Figure omitted). The correlation between OWI and  $SC_{WS}$  was significant (-0.68, above the 99% 204 confidence level) during P1 and became insignificant (0.20) during P2 (Figure 3c). Oppositely, the correlation 205 with SIGR enhanced from 0.4 in P1 to 0.62 in P2 (Figure 3d). Interestingly, the connections between these two 206 preceding factors and the EU pattern illustrated similar decadal changes (Figure 3 c, d). That is, the correlation 207 between EU and  $SC_{WS}$  was only significant (-0.62) in the former period; however, the correlation between EU 208 and SIGR was only significant (0.61) after the mid-1990s (Figure 3 c, d). Furthermore, the SIGR and SCWS were 209 mutually independent because the 21-year running correlation coefficient between them was maintained at a low 210 level (Figure S7). Therefore, we speculated that the impacts of the summer EU pattern on ground-level  $O_3$ 211 pollution in North China were robust and long-standing (Figure 1b). However, changed from SC<sub>WS</sub> in P1 to SI<sub>GR</sub> 212 in P2 (Figure 3 c, d).

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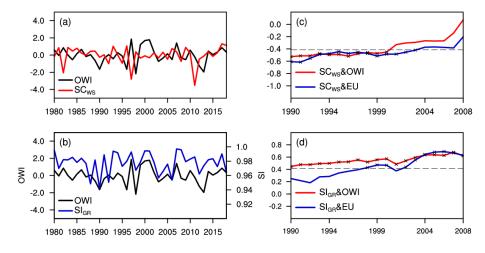


Figure 3. The normalized variation in (a) OWI (black) and  $SC_{WS}$  (red), (b) OWI (black) and  $SI_{GR}$  (blue) from 1980 to 2018 after detrending. The 21-year sliding correlation coefficients between (c)  $SC_{WS}$  and OWI (red), EU (blue), (d)  $SI_{GR}$  and OWI (red), EU (blue). The black dotted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

# 217 4. Possible physical mechanisms

218 The physical mechanisms how to achieve the impacts of SC<sub>WS</sub> on surface O<sub>3</sub> pollution in North China is still 219 a new question to the best of our knowledge. As an efficient climate forcing, the snow cover anomalies could 220 stimulate synchronous responses in the atmosphere by changing albedo and hydrological effects and could then 221 impact the atmosphere in the following seasons (Cohen and Rind, 1991). In April and May, the snow at high 222 latitudes began to melt and had obvious interannual variations, as shown by both the observations and the 223 reanalysis data (Figure S5). Generally, lower albedo, associated with less snow cover, meant that the land/snow 224 surface reflected less solar radiation and resulted in higher SAT. Warmer surfaces produce stronger longwave 225 radiation and heat the local atmosphere from the surface to the mid-troposphere (Chen et al., 2003; Chen et al., 226 2016). Moreover, the changing local soil moisture enhanced the surface heat flux and thus resulted in higher SAT 227 and atmospheric temperatures (Zhang et al., 2017). Finally, the warmed thermal conditions in the atmosphere 228 enhanced the local 1000-500 hPa thickness and represented positive anomalies of Z500 (Chen et al., 2003; Halder 229 and Dirmeyer, 2017). Compared to P1, the south edge of the area with high concentrations of snow (>85%) in late 230 spring shifted northward by approximately 2° in latitude during P2 (Figure 4a). Similarly, the significant changes 231 in radiation flux (shortwave + longwave) and heat flux (latent + sensible) also moved northward in P2 relative to 232 P1 (Figure 4 b, c). We speculated that this northward movement of effective snow cover, accompanied by shifts 233 in net heat flux, possibly contributed to the changing relationship between the SC<sub>WS</sub> and OWI.





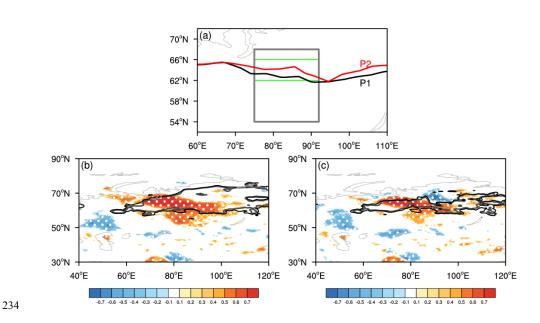


Figure 4. (a) The south edge of the 85% snow cover concontration during 1980–1993 (black) and during 2004–2018 (red). The gray (green) box represents the key area used to calculate the NHF<sub>WS</sub> (SCsw) index. The correlation coefficients between the SC<sub>WS</sub>× -1 and (b) surface net radiation 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.

241 The local responses of geopotential height in the mid-troposphere induced by negative anomalies of the SC<sub>WS</sub> illustrated decadal changes; that is, the significant correlation coefficients between SC<sub>WS</sub>  $\times$  -1 and Z500, as well 242 243 as SAT, were distributed more southward and were stronger in P1 (Figure 5a) than in P2 (Figure 5c). For 244 convenience, the roles of radiation and heat flux (shortwave + longwave + latent + sensible) were considered 245 together as net heat flux (Zhang et al., 2017), which was averaged over West Siberia (54°-68°N, 75°-92°E) and 246 defined as NHF<sub>WS</sub>. It was evident that the atmospheric responses associated with the NHF<sub>WS</sub> agreed well with 247 those of less SC<sub>WS</sub> (Figure 5 b, d). That is, the enhanced net heat flux related to decreased snow cover in West 248 Siberia heated the above atmosphere and resulted in local warmer SAT and anticyclonic circulations in the mid-249 troposphere during P1 (Figure 5 a, b). In addition, cyclonic responses can be found on the left and right sides of 250 the aforementioned anticyclonic anomalies in April-May (Figure 5 a, b). However, similar to the radiation and 251 heat flux in Figure 4 b-c, the atmospheric responses were distributed more northward and were weaker during P2 252 than during P1 (Figure 5 c, d).





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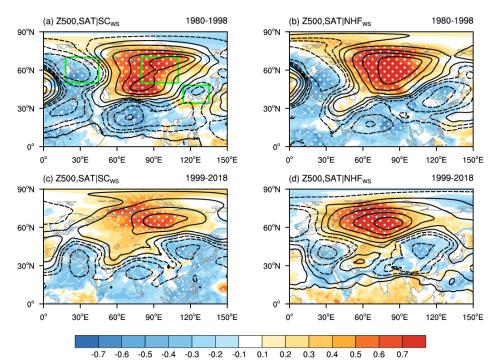
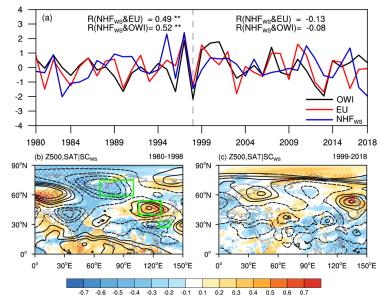


Figure 5. The correlation coefficients between the  $SC_{WS} \sim -1$  (a, c), NHF<sub>WS</sub> (b, d) and surface air temperature (shading) and geopotential height at 500 hPa (contour) from 1980 to 1998 (a, b) and from 1999 to 2018 (c, d). The white dots (hatching) indicate that the correlation coefficients 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.

258 The AM-mean NHF<sub>WS</sub> showed significantly positive correlations with both the summer-mean EU (0.49) and 259 OWI (0.52) during P1 (Figure 6a, S8a). Furthermore, the "-+-" anomalous atmospheric centers in April-May 260 (green boxes in Figure 5a) had significantly positive correlations with the summer EU pattern (CC=0.45, above 261 the 95% confidence level). The atmospheric anomalies stimulated by negative SC<sub>WS</sub> could appear as positive 262 phases of the EU pattern in JJA during P1 (Figure 6b, S8a). As one center of the EU pattern, the anticyclonic 263 anomalies over North China were significant in the mid- and lower-troposphere (Figure 6b, 7a) and resulted in 264 clear skies (Figure 7c). Sinking heating, intense sunlight (Figure 7c) and less precipitation (Figure 7a) resulted in 265 beneficial environments for the natural emissions of O<sub>3</sub> precursors (Lu et al., 2019) and photochemical reactions 266 (Pu et al., 2017). Differently, the northward and weaker atmospheric responses in April-May were almost 267 dispersed in summer (Figure 6c, S8b) and had little impacts on the local OWI in North China (Figure 7 b, d) 268 during P2, which were consistent with the insignificant correlations between the NHF<sub>WS</sub> and the EU (OWI) 269 (Figure 6a).

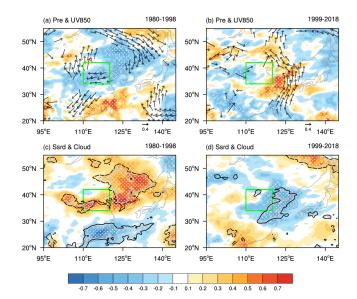






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Figure 6. (a) The normalized variation in the JJA OWI (black), JJA EU index (red) and AM NHF<sub>WS</sub> (blue) from 1980 to 2018 after detrending. The numbers represent the correlation coefficients between the NHF<sub>WS</sub> and EU, OWI during 1980–1998 and 1999–2018, respectively. Two asterisks indicate that the correlation coefficients exceeded the 95% confidence level. The correlation coefficients between SC<sub>WS</sub>× -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.



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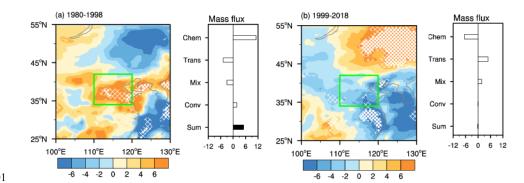
Figure 7. The meteorological conditions associated with the  $SC_{WS} \times -1$ . (a, b) The correlation coefficients between  $SC_{WS} \times -1$ and precipitation (shading) and wind at 850 hPa (arrow); (c, d) surface incoming shortwave flux (shading), and the sum of low and medium cloud cover (contour)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

285 In the  $EX_{SC}$  experiment, the simulated MDA8 O<sub>3</sub> and mass fluxes of ozone were composited (three lowest SC<sub>WS</sub> minus highest) during P1 and P2, respectively. During P1, the composited results (with fixed emissions) 286 287 were significantly positive (Figure 8a) and were in good agreement with the proposed mechanisms (i.e., less snow 288 cover in West Siberia resulted in severe surface O3 pollution in North China). The responses of MDA8 O3 pollution 289 in North China were insignificant during P2 (Figure 8b) and were also consistent with both weak impacts in this 290 period and changing relationships. Mass balance of ozone are jointly determined by four processes (i.e., chemistry, 291 transport, PBL mixing and convection) which could be isolated by the GEOS-Chem model. During P1, the 292 composite results of chemical reaction had large positive values (11.05 Tons  $d^{-1}$ ) (Figure 8a), indicating that the 293 dry-hot meteorological conditions were conductive to produce more O<sub>3</sub>. Anomalous anticyclonic circulations 294 located above the North China region resulted in downward air flow that may bring the ozone from the 295 stratosphere to surface. Hence, the value of convection was also positive. The values of transport and mixing were 296 negative (Figure 8a), but the sum of all processes was positive, indicating the ozone concentrations in North China 297 would increase. However, the composite results of chemical, transport and mixing were opposite (Figure 8b) 298 during P2 compared with P1. Meanwhile, the values of convection and the sum were extremely close to zero (Figure 8b), indicating that there were little impacts on ozone in North China when the SC<sub>WS</sub> was low during P2. 299 300 The composite results of mass fluxes were well agreement with the previous conclusion.





**Figure 8.** Composite difference of the summer MDA8 O<sub>3</sub> (unit:  $\mu$ g m<sup>-3</sup>) simulated by the GEOS-Chem model between the three lowest and the three highest SC<sub>WS</sub> years (a) from 1980 to 1998 and (b) from 1999 to 2018. The 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 mass fluxes of ozone (unit: tons d<sup>-1</sup>) during each periods. The left axis is the name of the physical-chemical processes: chemical reaction (Chem), transport (Trans), PBL mixing (Mix), convection (Conv) and their sums (Sum). The results were calculated within the planetary boundary layer.



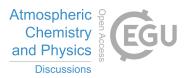


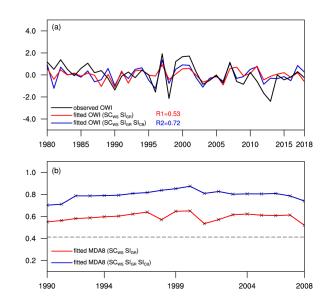
## 308 5. Conclusions and discussion

309 In this study, the April-May snow cover in West Siberia was newly proposed as a preceding climate driver 310 that influenced the surface O<sub>3</sub>-related meteorology in North China during 1980–1998, and the associated physical 311 mechanisms were also explained by comparing the periods before and after the mid-1990s. Accompanying the 312 northward shift of dense snow cover, the associated radiation and heat flux also retreated toward the polar region 313 during 1999-2018 (Figure 4); thus, the induced atmospheric anomalies were located northward in April-May and 314 disappeared in summer (Figure 5 c, d). However, in the period of 1980-1998, the positive phase of the EU pattern 315 in summer could be stimulated by negative anomalies of snow cover (mainly by enhanced net heat flux) in West 316 Siberia (Figure 6). Consequently, hot-dry air and intense solar radiation under anomalous anticyclonic circulations 317 not only enhanced the natural emissions of O<sub>3</sub> precursors but also promoted photochemical reactions to produce 318 more O<sub>3</sub> near the surface (Figure 7, 8). To enhance the robustness of this study, the ERA5 reanalysis data were 319 also employed to reproduce the observational analyses. As shown in Figure S9, identical results were obtained 320 and confirmed.

321 The linkage between the EU pattern and MDA8 O<sub>3</sub> was robust, which bridged the SC<sub>WS</sub> and OWI in the 322 period of 1980–1998 but connected the SI<sub>GR</sub> and OWI after the mid-1990s. In Figure 9, the OWI were regressed 323 by SC<sub>WS</sub> and SI<sub>GR</sub> from 1980–2018. The 21-year running correlation coefficient between the OWI and the fitted 324 values stably maintained around 0.6 and indicated that these two preceding factors almost introduced the full 325 impacts of the EU pattern (Figure 1b) over the whole period. Generally, the decadal changes in the climate drivers 326 influences the stability of the predictability. It is evident that our results overcame this problem and deepened the 327 understanding of variations in summer O<sub>3</sub> from the climate perspective. Yin et al. (2020a) also found that the sea 328 ice anomalies over the Canada Basin and the Beaufort Sea (Figure S1) also stimulated a Rossby-wave-like train 329 propagating through the North Pacific to influence the variability in the OWI in North China. When we added 330 these sea ice anomalies into the regressions, the fitting performance was visibly improved because the 21-year 331 running correlation coefficient was elevated to approximately 0.8 with OWI, as seen in Figure 9b.







332

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.

337 The concentrations of surface O<sub>3</sub> have been extensively measured since 2014 in China; this time scale cannot 338 support the study of the interannual-decadal variability in O3 pollution. In this study, we used two datasets, i.e., 339 the ozone weather index and the O3 concentrations simulated by GEOS-Chem, to focus on the impacts of climate 340 variability on surface O<sub>3</sub> in North China. Although the feasibility of these datasets was strictly examined, there 341 were still gaps between the real variations in O<sub>3</sub> and the variations in these two substitutions; this discrepancy 342 requires further research. Furthermore, there is no doubt that anthropogenic emissions are the fundamental drivers 343 of O<sub>3</sub> pollution, which has been investigated in many previous studies (Li et al., 2018; Li et al., 2019; Dang et al., 344 2020). After removal of the linear trend, the signals of climate warming in the atmosphere were also eliminated, 345 which allowed us to focus on the interannual variations. In addition, the decrease in haze aerosols was also proven 346 to be an effective contributor to recent interannual variations in O<sub>3</sub> concentrations (Li et al., 2019), which were 347 not involved in our study and need further attentions.





348 Data Availability. Hourly O3 concentration data can be downloaded from https://quotsoft.net/air/ (Ministry of 349 Environmental Protection of China, last accessed on 8 November 2020). Sea ice concentration data are from 350 https://www.metoffice.gov.uk/hadobs/hadisst/data/download.html (Met Office Hadley Centre, 2020). Snow cover 351 data can be downloaded from Rutgers University at http://climate.rutgers.edu/snowcover/ (Rutgers University, 352 2020). The observed snow data from meteorological stations are available at http://meteo.ru/tech/aisori.php. The 353 monthly mean MERRA2 reanalysis datasets are available at https://disc.gsfc.nasa.gov/datasets?page=1 (last 354 access: 21 March 2021). The monthly mean ERA5 reanalysis datasets are available at 355 https://cds.climate.copernicus.eu/cdsapp#!/home (Copernicus Climate Change Service, last accessed on 9 356 November 2020).

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# 360 Authors' contribution

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

# 363 **Competing interests**

364 The authors declare no conflict of interest.





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#### 469 Table and Figure captions

470 Figure 1. (a) The normalized variation in JJA-mean OWI (black), EU index (blue), simulated MDA8 O<sub>3</sub> (red) 471 from 1980 to 2018 and observed MDA8 O<sub>3</sub> (green) from 2014 to 2018 after detrending. (b) The 21-year sliding 472 correlation coefficients between simulated MDA8 O<sub>3</sub> (red), OWI (black) and EU. The black dotted line (crosses) 473 indicates (exceeded) the 95% confidence level. (c) Composite difference of the simulated MDA8 O<sub>3</sub> (unit: µg m<sup>-3</sup>) 474 in summer between the six highest and the six lowest EU index years from 1980 to 2018. The white dots (hatching) 475 indicate that the difference was above the 95% (90%) confidence level (t test). The green box represents the 476 location of North China. (d) Composite difference of the mass fluxes of summer ozone (unit: tons  $d^{-1}$ ) from the 477 GEOS-Chem between the six highest and the six lowest EU years from 1980 to 2018. The left axis is the name of 478 the physical-chemical processes: chemical reaction (Chem), transport (Trans), PBL mixing (Mix), convection 479 (Conv) and their sums (Sum).

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.

Figure 3. The normalized variation in (a) OWI (black) and SC<sub>WS</sub> (red), (b) OWI (black) and SI<sub>GR</sub> (blue) from
1980 to 2018 after detrending. The 21-year sliding correlation coefficients between (c) SC<sub>WS</sub> and OWI (red), EU
(blue), (d) SI<sub>GR</sub> 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<sub>WS</sub> (SCsw) index. The correlation coefficients between the SC<sub>WS</sub>× -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.

Figure 5. The correlation coefficients between the  $SC_{WS} \times -1$  (a, c), NHF<sub>WS</sub> (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.

Figure 6. (a) The normalized variation in the JJA OWI (black), JJA EU index (red) and AM NHF<sub>ws</sub> (blue) from 1980 to 2018 after detrending. The numbers represent the correlation coefficients between the NHF<sub>ws</sub> 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





- 507 linear trend is removed.
- 508 Figure 7. The meteorological conditions associated with the  $SC_{WS} \times -1$ . (a, b) The correlation coefficients 509 between SC<sub>WS</sub>× -1 and precipitation (shading) and wind at 850 hPa (arrow); (c, d) surface incoming shortwave 510 flux (shading), and the sum of low and medium cloud cover (contour) from 1980 to 1998 (a, c) and from 1999 to 511 2018 (b, d). The white dots (hatching) indicate that the correlation coefficients represented with shading exceeded 512 the 95% (90%) confidence level (t test). The gray (black) contours exceeded the 95% (90%) confidence level. The 513 green boxes represent the location of North China. The linear trend is removed. 514 Figure 8. Composite difference of the summer MDA8 O<sub>3</sub> (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 515 516 white dots (hatching) indicate that the difference was above the 95% (90%) confidence level (t test). The green 517 boxes represent the location of North China. The bar chart on the right is the composite difference of the summer 518 mass fluxes of ozone (unit: tons d-1) during each periods. The left axis is the name of the physical-chemical 519 processes: chemical reaction (Chem), transport (Trans), PBL mixing (Mix), convection (Conv) and their sums 520 (Sum). The results were calculated within the planetary boundary layer. 521 Figure 9. (a) The variation in the JJA-mean observed OWI (black), the fitted OWI-1 (by the SC<sub>WS</sub> and SI<sub>GR</sub>, red), 522 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
- 524 dotted line (crosses) indicates (exceeded) the 95% confidence level.