1	Intensified modulation of <u>winter</u> aerosol pollution in China
2	by El Niño with short duration
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22 Abstract

23 El Niño-Southern Oscillation (ENSO), a phenomenon of periodic changes in sea 24 surface temperature in the equatorial central eastern Pacific Ocean, is the strongest signal of interannual variability in the climate system with a quasi-period of 2-7 years. 25 El Niño events have been shown to have important influences on meteorological 26 27 conditions in China. In this study, the impacts of El Niño with different durations on 28 aerosol concentrations and haze days during December-January-February (DJF) in 29 China are quantitatively examined using the state-of-the-science Energy Exascale Earth System Model version 1 (E3SMv1). We find that $PM_{2.5}$ concentrations are 30 31 increased by 1-2 µg m⁻³ in the northeastern and southern China and decreased by up to 2.4 µg m⁻³ in central-eastern China during El Niño events relative to the 32 33 climatological means. Compared to long duration (LD) El Niño events, El Niño with 34 short duration (SD) but strong intensity causes northerly wind anomalies over 35 central-eastern China, which is favorable for aerosol dispersion over this region. Moreover, the anomalous southeasterly winds weaken the wintertime prevailing 36 37 northwesterly in northeastern China and facilitate aerosol transport from South and 38 Southeast Asia, enhancing aerosol increase in northeastern China during SD El Niño events relative to LD El Niño events. In addition, the modulation on haze days by SD 39 El Niño events is 2-3 times more than that by LD El Niño events in China. The 40 aerosol variations during El Niño events are mainly controlled by anomalous aerosol 41 42 accumulation/dispersion and transport due to changes in atmospheric circulation, while El Niño-induced precipitation change has little effect. The occurrence frequency 43 44 of SD El Niño events has been increasing significantly in recent decades, especially after 1940s, suggesting that El Niño with short duration has exerted increasingly 45 删除[z]: have intense modulation on aerosol pollution in China over the past few decades. 46

48 **1. Introduction**

49 Since the beginning of the 21^{st} century, China has experienced frequent events of 50 heavy haze pollution (Yang et al., 2018). The excessive aerosol concentrations during the heavy haze events can cause a large decrease in atmospheric visibility (Han et al., 51 2013) and pose significant public health hazards, such as a dramatic increase in 52 53 cardiovascular and respiratory diseases and associated mortality rates (Liu et al., 2019). PM_{2.5} (particulate matter less than 2.5 µm in diameter) has been reported to be 54 the fifth leading risk factor for mortality, inducing 7.6% of total deaths globally in 55 2015 (Cohen et al., 2017). In order to alleviate air pollution, a comprehensive and 56 57 better scientific understanding of factors that can affect aerosol concentrations and haze pollution in China is required. 58

59 Undoubtedly, the rise of anthropogenic emissions is the fundamental reason for the increase in aerosol concentration and haze pollution events (Huang et al., 2014), 60 61 but the unfavorable meteorological condition, as one of the most important external 62 factors, has been reported to have substantial influences on haze formation (Yang et 63 al., 2016a; Wang et al., 2019, 2020a). With increasing greenhouse gases in the future (2050-2099), severe winter haze events in Beijing would become 50% more frequent 64 and 80% longer in duration, compared to the historical period (1950-1999), due to an 65 accelerated warming of the lower atmosphere and weakening of the East Asian winter 66 monsoon (Cai et al. ,2017). In addition, external forcings, such as Pacific Decadal 67 68 Oscillation (Zhao et al., 2016) and Arctic sea ice (Wang et al., 2015; Zou et al., 2020), all have important impacts on aerosol concentrations and haze pollution in China. El 69 Niño-Southern Oscillation (ENSO), as another prominent climate phenomenon 70 71 caused by the coupled atmosphere-ocean interactions in the tropical Pacific Ocean 72 (Trenberth, 2019), is a significant signal of interannual climate change on a global scale. It triggers atmospheric circulation and precipitation anomalies globally (Yang et 73 74 al., 2016b, 2016c) and certainly has an important impact on haze events and aerosol 75 concentrations in China by modulating the East Asian winter monsoon system (Sakai 76 and Kawamura, 2009; Wang et al., 2000; Zhang et al., 2017).

The ENSO cycle is composed of warm-phase (i.e., El Niño) and cool-phase (i.e., La Niña) of sea surface temperatures (SSTs) over the tropical eastern Pacific Ocean, which further cause precipitation, atmospheric circulation and temperature anomalies in much of the tropics and subtropics. Such changes also affect the spatiotemporal 删除[z]: China has been continuously taking clean air actions in the recent years to battle for the blue sky. If all goes as planned, by 2035, the quality of atmospheric environment will be fundamentally improved and the goal of a beautiful China will be basically achieved. However, this goal requires

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81	distribution of aerosols in China (Feng et al., 2017, 2020; Sun et al., 2018; Yang et al.,
82	2014; Zhao et al., 2018; Zhu et al., 2012; Wang et al., 2020b). During a strong El
83	Niño event in 2015/2016, PM _{2.5} concentrations in winter were observed to increase by
84	20-100 μ g/m ³ in eastern China compared to that in 2014, which was attributed to the
85	weakened wind speed in the North China Plain during the El Niño event (Chang et al.,
86	2016; Wang et al., 2020a). $PM_{2.5}$ concentrations in southern China were also
87	decreased by about 20 $\mu g/m^3$ during the 2015/2016 El Niño event, which was
88	attributed to an enhanced precipitation and aerosol wet scavenging over this region.
89	Many studies counted haze days based on atmospheric visibility and found that El
90	Niño events could induce more (fewer) winter haze days in northern (southern) China
91	(Gao and Li, 2015; Li et al., 2017). In addition to surface observations, several studies
92	have also analyzed the relationship between ENSO events and aerosol loading based
93	on aerosol optical depth (AOD) data from satellite retrievals, (Jeoung et al., 2014; Sun
94	et al., 2018). Jeoung et al. (2014) analyzed the combined AOD data of MODIS
95	(Moderate Resolution Imaging Spectroradiometer), MISR (Multi-angle Imaging
96	SpectroRadiometer) and AERONET (Aerosol Robotic Network) and found that
97	during the warm phase of ENSO, the fine-mode AOD increased in eastern coastal
98	areas but decreased in some inland areas of China. Sun et al. (2018) studied the
99	influence of ENSO events on the interannual variation of wintertime aerosol in China
100	using AOD data (1980-2016) from MERRA-2 reanalysis and found that AOD in the
101	North China Plain increased significantly during El Niño events, with a 15%
102	increment in the Beijing-Tianjin-Hebei region compared to the long-term average.
103	They also pointed out that AOD increased in eastern and southern China and
104	decreased in southwestern China during El Niño events.
105	Although observational data showed that aerosols in China were largely

106 perturbed during El Niño events, the individual impacts of atmospheric circulation 107 and precipitation anomalies associated with El Niño could not be simply extracted out 108 with observations alone. Numerical simulations have been used to isolate the individual impacts of El Niño on aerosols in China through a superposed SST 109 perturbation method and explore the underlying mechanisms (Yu et al., 2019; Zhao et 110 111 al., 2018). Based on an aerosol-climate coupling model, Zhao et al. (2018) suggested that El Niño increased the seasonal mean aerosol concentration in southern China in 112 113 winter, which is mainly due to the increased aerosol transport from South and Southeast Asia. Using the same model, Yu et al. (2019) showed that, relative to the 114

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climatological mean, wintertime surface aerosol concentrations in northeastern and 115 southeastern China (central and southwestern China) increased (decreased) during El 116 117 Niño events, which was mainly attributed to anomalies in near-surface winds and the 118 resulting aerosol mass flux divergences. Sun et al. (2018) used the aerosol-climate model CAM5 to simulate the impact of ENSO events on the interannual variability of 119 120 AOD in China and found that El Niño events led to an increase in AOD in central and 121 eastern China. They suggested that the change in AOD was mainly dominated by the 122 change in meridional winds.

123 Some studies focused on the effects of different spatial types (e.g., East Pacific and Central Pacific El Niño, Kao and Yu (2009)) and intensities of El Niño events on 124 125 aerosol concentrations in China (e.g., Yu et al., 2019). Yu et al. (2019) found that, due 126 to the difference in atmospheric circulation between two types of El Niño, Central Pacific El Niño events resulted in a larger increase in aerosol burden in southern 127 China than East Pacific El Niño events. They also indicated that a moderate El Niño 128 129 event led to an increase in seasonal mean near-surface aerosol concentrations 130 throughout eastern China in winter, while a strong or weak El Niño event brought 131 about a significant decrease in aerosol concentrations in northern China.

132 Apart from the spatial types with different intensities, El Niño can also be 133 categorized as short duration (SD) and long duration (LD) according to the length of 134 their decay period (Boo et al., 2004; Chen et al., 2012; Guo and Tan, 2018). These two 135 temporal types of El Niño events have been confirmed to have different impacts on 136 the SSTs, vertical wind shear, relative humidity and precipitation in South China Sea and Philippine Sea (Guo and Tan, 2018; Wu et al., 2019). The El Niño events with 137 138 different durations are likely to have different impacts on the aerosol distribution in China. However, few studies explore the different impacts of SD and LD El Niño 139 140 events on aerosol concentrations and haze days in China, as well as the associated mechanisms, which are essential for air pollution control in the near future. 141

In this study, the effects of SD and LD El Niño events on wintertime aerosols in China are investigated by using the state-of-the-science Energy Exascale Earth System Model (E3SM). The data, model, and analysis methods used in this research are presented in Section 2. The influences of different durations of El Niño events on aerosols over China and the mechanisms involved are analyzed in Section 3. Summary of the main results and discussion of the implications for future research are provided in Section 4.

2. Data and Methods 149

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150	2.1 Data
151	We use the following datasets in this study.
152	(1) The merged Hadley-NOAA/OI SST and sea ice concentration (SIC) datasets
153	from 1870 to 2017 with a horizontal resolution of $1^{\circ} \times 1^{\circ}$ (Hurrell et al., 2008)
154	are used to obtain the climatological mean SST and SIC pattern and the
155	anomalies of SST during SD and LD El Niño events.
156	(2) Monthly mean emissions of aerosols and their precursors in 2014 from the
157	CMIP6 (Coupled Model Intercomparison Project Phase 6) (Hoesly et al.,
158	2018; van Marle et al., 2017) with emissions in China replaced by MEIC
159	(multi-resolution emission inventory for China) emission inventory are used
160	as input datasets in model simulations.
161	(3) Hourly observations of PM _{2.5} concentrations at 1657 stations over China
162	from December 2014 to February 2015 derived from the China National
163	Environmental Monitoring Centre (CNEMC) are applied to evaluate the 删除[z]: (http://www.cnemc.cn)
164	model performance.
165	(4) Monthly averaged ERA5 reanalysis data from 1950 to 2017 (Hersbach et al.,
166	2020) provided by European Centre for Medium-Range Weather Forecasts
167	(ECMWF) are used to evaluate the simulated meteorological parameters
168	during El Niño events.
169	2.2 SD and LD El Niño events
170	Here we first describe how the LD and SD El Niño events are defined. The year
171	in which El Niño developed is denoted by year ⁰ and the months of that year are

denoted by Jan⁰, Feb⁰, ..., and Dec⁰, while the following year and months are year¹

- and Jan¹, Feb¹,, and Dec¹, respectively. Niño 3.4 index is detrended SST anomaly 173
- over the Niño 3.4 region (170°W-120°W, 5°S-5°N). El Niño event is firstly identified 174
- when a 3-month running mean Niño 3.4 index, is greater than 0.75°C in any month 175
- from Oct⁰ to Feb¹ of its developing phase. If the Niño 3.4 index is higher than 0.5°C 176
- in any month from Oct¹ to Feb² of <u>its</u> decaying phase, the El Niño event is an LD El 177
- Niño event; otherwise, it is an SD El Niño event (Wu et al., 2019). 178

Figure 1 shows the time series of the Niño3.4 indices calculated based on the 179

- Hadley-NOAA/OI data for the period 1870-2017. According to the definition above, 180
- totally 30 El Niño events are identified in this time period, with 22 SD El Niño events 181

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删除[z]:, defined as the regional mean linear detrended SST anomaly from the monthly mean climatology over the Niño3.4 region (170°W-120°W, 5°S-5°N),

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182 (1877/1878, 1885/1886, 1888/1889, 1896/1897, 1902/1903, 1911/1912, 1923/1924,

- 183 1925/1926, 1930/1931, 1951/1952, 1957/1958, 1963/1964, 1965/1966, 1972/1973,
- 184 1982/1983, 1991/1992, 1994/1995, 1997/1998, 2002/2003, 2006/2007, 2009/2010,
- 185 2015/2016 as developing phase) and 8 LD El Niño events (1899/1900, 1904/1905,
- 186 1913/1914, 1918/1919, 1939/1940, 1968/1969, 1976/1977, 1986/1987 as developing
- 187 phase). The temporal evolution of the Niño3.4 indices during the developing and

188 decaying phases of SD and LD El Niño events is shown in Figure 2. During the

developing phase from Jul^0 to Feb¹, due to the fast accumulation of ocean heat content

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- and rapid adjustments of the surrounding seas to the tropical Pacific Ocean warming
- 191 (Wu et al., 2019), the Niño3.4 indices are higher in SD El Niño events, but the SST
- 192 <u>anomaly</u> decreases rapidly in the decaying phase, compared to those in LD events.

2.3 Model description and experimental design

194 To quantify the influence of El Niño with various durations on aerosols in China, the U.S. Department of Energy (DOE) E3SM version 1 (E3SMv1) is utilized in this 195 196 study, which includes atmosphere, land, ocean, sea ice and river components (Golaz et 197 al., 2019). It was branched from the CESM1 (Community Earth System Model) but 198 has been updated substantially since. The E3SM Atmosphere Model version 1 (EAMv1) is a descendant of the well-known CAM5.3 (Community Atmosphere 199 200 Model version 5.3) (Rasch et al., 2019). It includes considerable upgrades to aerosols, turbulence, chemistry and cloud related processes. EAMv1 provides various options 201 202 of spatial resolution. In this study, the horizontal spatial resolution of approximately 203 1° and 30 vertical layers from the surface to 3.6 hPa are used in the model configuration. The model simulates aerosols including sulfate, black carbon (BC), 204 primary organic aerosol (POA), secondary organic aerosols (SOA), sea salt, and 205 mineral dust in the four-mode Modal Aerosol Module (MAM4) (Wang et al., 2020). 206

207 The following numerical experiments are conducted in this study. A "CLIM" experiment is driven by climatological average of monthly SST and SIC over 208 209 1870-2017 and integrated for 20 years. Two sets of sensitivity experiments, "SD" and 210 "LD", are respectively driven by the monthly SST representing composite SD and LD El Niño events. The monthly SSTs representing SD (LD) El Niño events are produced 211 through superposing the average of monthly SSTs from Jul^0 to Jun^1 of the 22 SD (8) 212 LD) El Niño events selected in Sec. 2.2 on top of the climatological monthly SST 213 214 over 60°S-60°N. Each set of sensitivity experiment has 3 ensemble members with 215 different initial conditions branched from different years of the CLIM experiment.

216	Each member of the sensitivity experiments is run for 8 years with first 3 years used					
217	for spin-up and the last 5 years used for analysis. The differences in the monthly and					
218	daily mean model fields between SD, LD and CLIM are used to analyze the effects of					
219	duration of El Niño events on aerosols. To understand the potential mechanism of El					
220	Niño impacts on aerosol pollution in China, two additional experiments are also					
221	conducted. The "SD_emis" experiment is the same as the first ensemble member of					
222	SD experiment, except that the emissions of aerosols and precursor gases from South					
223	and Southeast Asia are turned off. The "CLIM_emis" experiment is same as the					
224	"SD_emis" experiment but driven by climatological average of monthly SST and SIC					
225	over 1870-2017. All other external forcings, including insolation, greenhouse gas					
226	concentrations, and emissions of aerosol and precursor are kept at present-day					
227	conditions (year 2014), In brief, the experiments performed are as follows.(Table 1). 删除[z]:					
228	1. CLIM: control simulation driven by climatological SST.					
229	2. SD: sensitivity simulation to quantify the impacts of El Niño events with					
230	short duration on aerosols in China. Same as CLIM except for the imposed					
231	SST pattern of short duration El Niño, (Fig. 3a). 删除[z]:					
232	3. LD: sensitivity simulation to quantify the impacts of El Niño events with					
233	long duration on aerosols in China. Same as CLIM except for the imposed					
234	SST pattern of long duration El Niño, (Fig. 3b). 删除[z]:					
235	4. SD_emis: sensitivity simulation to quantify the role of regional transport of					
236	aerosols from South and Southeast Asia on aerosols in China during El Niño					
237	events with short duration. Same as SD except that the emissions of aerosols					
238	and precursor gases from South and Southeast Asia are turned off.					
239	5. CLIM_emis: sensitivity simulation to serve as the baseline for SD_emis.					
240	Same as SD_emis except for the use of climatological SST.					
241	2.4 Model evaluation					
242	To evaluate the model performance in simulating aerosol concentration and					

distribution in China, the simulated December-January-February (DJF) mean surface PM_{2.5} (sum of sulfate, BC, POA and SOA in model simulation) concentrations from the CLIM experiment is compared with the observed PM_{2.5} concentrations in Fig. 4. The model well reproduced the spatial distribution of wintertime aerosols in China, with high aerosol concentrations in eastern China (e.g., North China Plain, Fenwei Plain and Yangtze River Delta) and southwestern China (e.g., Sichuan Basin) and low aerosol levels in western China. The spatial correlation coefficient (R) between the

- 250 E3SMv1 EAMv1 simulation and observations for near-surface PM_{2.5} concentrations is
- +0.43. However, the model underestimates the PM_{2.5} concentrations in China, with a 251
- 252 normalized mean bias (NMB) of -65.74% compared to the observed values, which 删除[z]: deviation
- 253 was also reported in many studies using the CESM1 model (e.g., Yang et al., 2017a,

b). The discrepancy could be due to many factors, including the lack of nitrate and 255 ammonium aerosols in the model, strong wet scavenging simulated at the mid- and 256 high latitudes, and less transformation from gas to particles. In addition, we focus on 257 anthropogenic aerosols. If natural dust is considered in the modeled $PM_{2.5}$ calculation, the NMB will drop to -6.38%. Nevertheless, the aerosol concentrations in EAMv1 258 simulations are closer to the observations than previous ENSO-aerosol studies (e.g., 259 260 Yu et al., 2019; Zhao et al., 2018) and the composite differences are analyzed in this

261 study rather than the climatological mean concentration. We don't expect the systematic low biases in PM_{2.5} concentrations affect our study on the impact of El 262 Niño events. However, we should note that the aerosol changes in China during 263

SD/LD El Niño events in the real world could be larger than the simulated values 264 265 here.

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3. Results 267

3.1 Impacts of SD and LD El Niño events on aerosol concentrations 268

269 Figure 5, show the absolute and relative impacts of the two types of El Niño events with different durations on the simulated DJF mean near-surface 270 concentrations and column burdens of PM_{2.5} in China. The effects of the SD and LD 271 272 El Niño events on near-surface aerosol concentrations over China are similar in the spatial pattern distribution, with increases in the northeastern and southern China by 273 about 1-2 µg m⁻³ (5-15% compared to the climatological mean) and decreases in 274 central-eastern China during El Niño events relative to the climatological averages. 275 276 This spatial pattern of aerosol changes is in accordance with previous modeling 277 studies (Feng et al., 2016; Yu et al., 2019; Zhao et al., 2018). However, the modeling 278 results are not exactly the same as the observed PM_{2.5} changes, which show increases in PM_{2.5} over northeastern China and the North China Plain and slightly decreases in 279 southern China during the 2015/2016 El Niño event (Chang et al., 2016). The 280 281 discrepancy between the model simulations and observations can be attributed to the following reasons. First of all, instead of the El Niño impacts, observed aerosols can 282

删除[z]: Figures 删除[z]: and 6 删除[z]:, respectively, be affected by other factors including East Asian winter monsoon (Yang et al., 2016a),
Arctic Oscillation (Zhang et al., 2019) and Pacific Decadal Oscillation (Zhao et al.,
2017), whereas the modeled changes are purely caused by the El Niño impacts
through the imposed SST perturbation. Secondly, the time coverage of near-surface
PM_{2.5} observations is limited in China and only one extreme El Niño event
(2015/2016) was analyzed in previous El Niño-PM_{2.5} studies (e.g., Chang et al., 2016),
which is not fully representative of the impact of general El Niño events.

290 Although the spatial patterns of the SD and LD El Niño influences on the DJF PM_{2.5} concentrations in China resemble each other, the magnitudes of the influences 291 292 are different. Central-eastern China experiences more reductions in near-surface PM_{2.5} 293 concentrations during SD El Niño, with the concentration decreases of more than 2.4 μg m⁻³ (15% relative to the climatological mean), which is much larger than the 0.6 294 μg m⁻³ (5%) during LD El Niño. In southern China, the spatial coverage of the 295 increase in PM_{2.5} concentration shrinks more during SD than LD El Niño events 296 297 relative to the CLIM, but the intensities of the anomalies triggered by the two 298 temporal types of El Niño events are similar. Moreover, SD El Niño induces a larger 299 increase in PM_{2.5} concentrations in northeastern China by 1.2 μ g m⁻³ (10%) than that of 0.6 µg m⁻³ (5%) during LD El Niño events. 300

301 The PM_{2.5} burden and near-surface concentration anomalies triggered by the El 302 Niño events with short and long durations are basically the same in spatial distribution 303 but with different magnitudes (Fig. 5). For example, the reduction in aerosol burden is much larger in central-eastern China during the SD El Niño events than during the LD 304 El Niño events, with maximum negative anomalies, respectively, reaching -1.6 and 305 306 -0.6 mg m⁻². Overall, SD El Niño events yield stronger impacts on aerosol pollution in China than LD El Niño events, especially in central-eastern China with negative 307 308 pollution anomalies.

309 3.2 Mechanisms of SD and LD El Niño impacts on aerosols

Since aerosols and their precursor gas emissions are prescribed at the same rates in the control (CLIM) and sensitivity (SD/LD) simulations, changes in meteorological factors such as circulation and precipitation play dominant roles in altering aerosol concentrations by influencing the regional transport and wet removal of aerosols (Yang et al., 2016a). Previous studies also suggested that aerosol variations during ENSO events were controlled by ENSO-related circulation and precipitation changes (Yu et al., 2019; Zhao et al., 2018). Here, we examine the atmospheric circulation and

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317 precipitation anomalies and the associated aerosol processes during the SD and LD El

318 Niño to explore the mechanisms of the two types of El Niño effects on aerosols in

319 China.

Both the SD and LD El Niño events trigger negative anomalies in sea level 320 321 pressure (SLP) over the eastern China and East China Sea and positive anomalies 322 over the Philippine Sea and Sea of Okhotsk (not shown), leading to anomalous 323 cyclonic and anticyclonic circulations over these regions, respectively (Figs. 6a and 删除[z]: 7a 324 (6b). At 850 hPa, the anomalous cyclonic circulation over the East China Sea causes 删除[z]:7b 325 anomalous northerly winds over central-eastern China, enhancing the prevailing northwesterly winds in winter. The enhanced winds favor the aerosol dispersion, 326 327 which explains the decrease in $PM_{2.5}$ concentrations over central-eastern China during 328 El Niño events relative to the climatological mean. In addition, at 500 hPa, most areas 329 over China have an anomalous low pressure (Figs. 6d and 6e), which increases the 删除[z]: 7d 330 atmospheric instability and strengthens the aerosol vertical mixing and dispersion 删除[z]: 7e 331 over central-eastern China. Over southern China, the aerosol variations are 332 significantly affected by the regional transport of particles from South and Southeast 333 Asia. During El Niño events, anomalous southwesterly winds at the northwest edge of the anomalous anticyclone over the Philippine Sea bring aerosols from South and 334 335 Southeast Asia to southern China, contributing to the aerosol increases in southern 336 China relative to the climatological mean (Figs. <u>6a</u> and <u>6b</u>). In the northeastern China, 删除[z]: 7a 337 anomalous southeasterly winds associated with the anomalous anticyclonic circulation 删除[z]:7b 338 over Sea of Okhotsk weaken the wintertime prevailing northwesterly winds, giving rise to the aerosol increases in the northeastern China during El Niño events. In 339 addition, the anomalous anticyclone brings aerosols from South and Southeast Asia to 340 northeastern China that will be discussed next, contributing to the aerosol pollution in 341 342 northeastern China during El Niño events. Compared to LD El Niño events, the negative anomaly of SLP over the East 343 344 China Sea during the SD El Niño events is stronger and extends deeply into the central-eastern China, resulting in anomalous northerly winds over central-eastern 345 346 China and southeasterly winds over northeastern China in the lower atmosphere (Fig. 347 <u>6c</u>). The wind anomalies intensify the aerosol dispersion in central-eastern and 删除[z]: 7c accumulation in northeastern China, leading to a stronger effect of El Niño with short 348 删除[z]: 7c 349 duration on the aerosol variation in China. Furthermore, the anomalous northerly winds in both lower atmosphere and 500 hPa over southern China (Figs. 6c and 6f) 350 删除[z]: 7f are unconducive to the regional transport of aerosols from South and Southeast Asia

to central-eastern China. 352 353 To further verify the model simulations in capturing atmospheric circulation anomalies during SD and LD El Niño events, the wind fields are compared with those 354 from ERA5 reanalysis data. The anomalous atmospheric circulation patterns in the 355 latest SD El Niño event (2015/2016) and LD El Niño event (1986/1987) relative to 356 357 the climatological mean (1950-2017) from the ERA5 are shown in Fig. 7. Overall, the 358 SD and LD El Niño-induced anomalous atmospheric circulations over China simulated in E3SM are in consistent with the reanalysis data. Both of them show the 359 anomalous northerly winds over central-eastern China at 850 hPa during SD El Niño 360 361 compare to LD El Niño. In addition, obvious anomalous cyclone at 500 hPa over most of China can be seen in both E3SM and ERA5. 362 In addition to the regional transport prompted by anomalous atmospheric 363 circulations, El Niño can influence aerosol wet removal through perturbing 364 precipitation. As described in Figs. 8a and 8b, the spatial patterns of winter 365 precipitation anomalies in China during SD and LD El Niño events are similar, with 366 367 positive anomalies located along the southeastern coastal areas due to the additional

moisture transport by anomalous southwesterly winds over the South China Sea.However, the two types of El Niño events differ in the magnitude of precipitation

anomalies. In central-eastern China, precipitation decreases during SD El Niño events,

371 compared to LD El Niño events, whereas precipitation increases over eastern coastal

areas and northeastern China (Fig. 8c). This is linked to the anomalous cyclonic

373 circulation over central-eastern China (Fig. <u>6c</u>), which hinders moisture from South
374 China Sea to central-eastern China but brings in moisture from Sea of Japan to

northeastern China. Over Pearl River Delta, precipitation decreases during SD El 375 376 Niño events but increases during LD El Niño events, which is also associated with the anomalous northerly winds and corresponding impact on moisture transport over this 377 region. In general, aerosol wet deposition decreases in central-eastern China and 378 379 increases over southern and northeastern China during El Niño events (Figs. 8d and 380 8e). With short duration but strong intensity, El Niño events have larger impacts on 381 aerosol wet removal than those with long duration (Fig. 8f). However, the wet removal shows a positive relationship with the aerosol concentration, which should be 382 383 a negative relationship in theory if other conditions remain unchanged. Water vapor

384 can accelerate the chemical transformation of secondary aerosols (Yang et al., 2015),

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391 Both the accumulation/dispersion of local aerosols within China and regional 392 transport of aerosols from South and Southeast Asia can contribute to the aerosol changes in China during El Niño events. With emissions of aerosols and precursor 393 394 gases in South and Southeast Asia turned off, the decrease pattern of PM_{2.5} over 9), 395 central-eastern China does not change (Fig. suggesting that accumulation/dispersion of local aerosols dominates the aerosol change over this 396 397 region during El Niño events. Over southern China, the increase of PM_{2.5} burden is 398 weakened when the South and Southeast Asian emissions are turned off, indicating 399 that regional transport of aerosols from South and Southeast Asia have a large 400 contribution to the aerosol variation over this region. It is interesting that, without 401 emissions from South and Southeast Asia, both near-surface concentration and column burden of PM_{2.5} in northeastern China decrease during El Niño events relative 402 403 to the climatological mean, but the change reverses to increase when the South and 404 Southeast Asian emissions are considered. It indicates that the aerosol enhancements 405 in northeastern China during the El Niño events are most likely influenced by aerosol 406 transport from South and Southeast Asia due to anomalous southeasterly winds at the 407 eastern edge of the anomalous cyclonic circulation in eastern China (Fig. 6c), which 408 warrants further analysis using a source-receptor model such as CAM5-EAST (Ren et al., 2020; Yang et al., 2020). 409

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410 **3.3 Quantitative impacts on regional PM_{2.5} concentrations and haze**

411 **days**

412	Figure 10 summarizes the simulated probability density distributions of PM _{2.5} 删除[z]: Table 1
413	concentrations, regional mean PM _{2.5} concentrations and number of haze days in DJF
414	over the sub-regions in China including the North China Plain (NCP, 35-41°N, 删除[z]: (Fig. 10),
415	114-120°E), Sichuan Basin (SCB, 28-33°N, 103-108°E), Yangtze River Delta (YRD,
416	29-34°N, 118-121.5°E), Pearl River Delta (PRD, 21.5-25°N, 111-116°E), Northeast
417	Plain (NEP, 41-48°N, 120-130°E), the Yunnan-Guizhou Plateau (YGP, 23-27°N,
418	100–110°E) and the Fenwei Plain (FWP 33–35°N 106–112°E and 35–38°N

419 110–114°E) from CLIM, SD and LD simulations. Haze days are defined as days with
420 daily near-surface PM_{2.5} concentrations above the 90th percentile of the CLIM PM_{2.5}

421 concentrations in each sub-region of China.

422 During El Niño events, DJF mean near-surface PM_{2.5} concentrations decrease 423 over NCP, SCB, YGP, and FWP regions and increase over PRD and NEP in both SD and LD, compared to CLIM. Although the $PM_{2.5}$ concentrations show an increase in 424 425 SD and a decrease in LD over YRD region, the changes are statistically insignificant 426 in this region (Fig. 5). SD El Niño events have a stronger modulation on aerosols in China than LD El Niño events. Over the regions with concentration decreases (NCP, 427 SCB, YGP, and FWP), regional mean near-surface PM_{2.5} concentration in LD is lower 428 than CLIM by 0.24 μ g m⁻³, while the reduction reaches 1.22 μ g m⁻³ in SD, about 5 429 times as that of LD. Over the regions with concentration increases (PRD and NEP), 430 the PM_{2.5} increase in SD relative to CLIM is 0.74 μ g m⁻³, which is also higher than 431 the 0.56 μ g m⁻³ in LD. 432 433 Similar to the PM_{2.5} concentration, the modulation of SD El Niño events on haze 434 days are 2-3 times as high as that of LD El Niño events. During LD El Niño events, 435 the number of haze days in DJF at NCP, SCB and FWP is reduced by 1.14, 0.73 and 1.53 days, respectively, compared to the climatological mean, while the decrease in 436 437 haze days during SD El Niño events is more substantial (1.87, 2.13 and 2.87 days). 438 The probability density distributions of $PM_{2.5}$ concentrations over NCP, SCB and 439 FWP in SD and LD also shift to the left, relative to CLIM (Figs. 10b, 10g, and 10h). Consistent with the stronger modulation of SD El Niño events discussed above, the 440 shift in SD is more than that in the LD simulation. In addition, YRD, PRD and NEP 441 442 regions all have increases in haze days in DJF during SD and LD, relative to CLIM. 443 Similarly, during SD and LD El Niño events, the probability density distributions of 444 high values of PM_{2.5} concentrations over YRD, PRD and NEP slightly shift to the right relative to CLIM, (Figs. 10c-e). The number of haze days in DJF over YGP 445 decreases during SD El Niño events by 1.4 days, but there is a slight increase of 0.4 446 days during LD El Niño events, likely due to the opposite aerosol changes in the 447 448 eastern and western parts of YGP region (Fig. 5). There are more (fewer) haze days in 449 both SD and LD than in CLIM over YRD, PRD and NEP (NCP, SCB and FWP), which is inconsistent with the simulated greater (less) precipitation over these regions 450 caused by El Niño events. It further indicates that anomalies in precipitation are not 451 452 the most dominant factor modulating winter haze days in China during El Niño events,

删除[z]: Fig. 11).

删除[z]: .

- 453 but rather the anomalous aerosol accumulation/dispersion and transport due to
- 454 anomalous atmospheric circulation.

455 **3.4 Historical increase in SD El Niño events**

456 Many studies have suggested an increase in the variability of El Niño events 457 under greenhouse warming (Cai et al., 2018; Grothe et al., 2020). However, few 458 studies have shown the historical changes in El Niño with different durations, which 459 would further impact aerosol concentrations and haze days in China.

Here we show the occurrence of SD and LD El Niño events since the 460 461 preindustrial era in Fig. <u>11</u>. The number of SD El Niño events fluctuated but has 462 increased significantly during the past few decades, especially after 1940s. The occurrence of SD El Niño increased from one event per fifteen years during 463 464 1941-1955 to four events per fifteen years during 2001-2015, with the increase at 465 confidence level of 92%, while LD El Niño events did not present a significant trend 466 in the historical period. Wu et al. (2019) found that the duration of El Niño is mainly influenced by the timing of onset, associated with the early onset of delayed negative 467 oceanic feedback as well as the fast adjustments of the tropical Indian and Atlantic 468 469 Oceans to the tropical Pacific Ocean warming. It is conjectured that the onset timing 470 of El Niño events gets earlier under greenhouse forcing, but the detailed analysis is 471 out of the scope of this study. Nevertheless, because the frequency of the El Niño events with short duration increased significantly, the modulation by El Niño events 472 473 on wintertime aerosols in China has intensified in the past few decades.

474

475 **4. Conclusion and discussions**

As a prominent climate phenomenon, El Niño triggers atmospheric circulation and precipitation anomalies on a global scale, thus having important effects on haze days and aerosol pollution in China. In this study, the impacts of different temporal types of El Niño events with short and long duration on aerosols in China are simulated using the state-of-the-science E3SM model.

For both SD and LD El Niño events, their changes to the DJF mean $PM_{2.5}$ concentrations have similar spatial distributions over China, relative to the climatological mean. The anomalous anticyclonic circulation over the Sea of Okhotsk weakens the prevailing northwesterly winds in DJF in northeastern China and enhances the accumulation of locally emitted aerosols, along with the anomalous 删除[z]: 12

486 southeasterly winds at the eastern edge of the anomalous cyclonic circulation in eastern China that intensifies the aerosol transport from South and Southeast Asia to 487 488 northeastern China. The near-surface PM_{2.5} concentration in northeastern China increases by 1-2 μ g m⁻³ during El Niño events relative to the climatological conditions. 489 In southern China, the anomalous anticyclonic circulation over the Philippine Sea 490 491 facilitates the transport of aerosols from South and Southeast Asia to southern China 492 and thus the near-surface $PM_{2.5}$ concentrations in southern China increase by 1-2 μ g 493 m⁻³. The decrease in near-surface PM_{2.5} concentrations in central-eastern China is mainly controlled by the enhanced northerly winds from the anomalous cyclonic 494 circulation over eastern China and the East China Sea, leading to the dispersion of 495 496 local aerosols, while precipitation change has little effect on aerosols here. Compared to LD El Niño events, due to the anomalous cyclonic circulation over eastern China, 497 SD El Niño events exhibit a stronger reduction (1-2 μ g m⁻³) in near-surface PM_{2.5} 498 concentrations over central-eastern China and a larger increase (0.6 μg m⁻³) in 499 500 northeastern China. Overall, El Niño with short duration has a stronger modulation on 501 wintertime aerosols in China than El Niño with long duration.

502 Compared with CLIM, mean near-surface $PM_{2.5}$ concentrations in DJF decrease 503 over NCP, SCB, <u>YGP</u> and FWP regions and increase over PRD and NEP in both SD 504 and LD, but the decrease over these regions in SD El Niño events reaches 1.22 µg m⁻³, 505 about 5 times as large as that of LD. Similarly, both SD and LD El Niño events induce 506 less (more) haze days in DJF than CLIM over NCP, SCB and FWP (YRD, PRD and 507 NEP). However, the decreases in haze days in DJF at NCP, SCB and FWP during SD 508 El Niño events are 2-3 times more than that during LD El Niño events.

509 We also found that the occurrence frequency of SD El Niño events increased from one event per fifteen years during 1941–1955 to four events per fifteen years 510 511 during 2001–2015, whereas LD El Niño events did not exhibit a significant trend in the historical period. In particular, seven SD El Niño events have occurred since the 512 1990s, but no LD El Niño event occurred. Compared to LD El Niño events, SD El 513 Niño events have a greater impact on wintertime aerosols over China. Therefore, the 514 515 impact of El Niño events on wintertime aerosols in China has intensified in the past 516 few decades due to their short durations.

517 Our results of the important effect of SD El Niño events and its recent 518 intensification are of great significance for the understanding of El Niño on China's 519 haze pollution, alleviating air pollution, and coping with climate change. The 删除[z]: YKP

520 simulated spatial patterns of aerosol changes during El Niño events resemble those in 521 previous studies (Feng et al., 2016; Yu et al., 2019; Zhao et al., 2018). However, there 522 are still some inadequacies remaining to be improved. Natural aerosols including dust and sea salt were not considered in this study. The EAMv1 model largely 523 underestimated PM2.5 concentration in China related to the lack of nitrate and 524 ammonium aerosols and other model biases. We also found that, during El Niño 525 526 events, more aerosols from South and Southeast Asia can be transported to 527 northeastern China, leading to an increase in aerosol concentrations over there. Thus, more in-depth analysis is needed in future studies. In addition, during the cooling 528 529 phase of ENSO, La Niña events may also have various durations and can have different impacts on air pollutions in China, which merits further investigation. Since 530 that $PM_{2.5}$ is more harmful to human health than PM_{10} , in this study, we focused on 531 PM_{2.5} rather than PM₁₀, which is largely contributed by natural dust aerosol. The 532 impacts of El Niño on dust will be investigated in our future work. 533

535 Data availability

- 536 The E3SMv1 model is available at https://e3sm.org/ (last access: <u>25 May</u> 2021). Our 删除[z]: 1 February
- 537 results can be made available upon request. Hourly observations of PM_{2.5}
- 538 concentrations over China can be derived from the China National Environmental
- 539 Monitoring Centre (http://www.cnemc.cn, last access: <u>25 May 2021). Monthly ERA5</u> 删除[z]: 1 February
- 540 <u>reanalysis data can be downloaded at</u>
- 541 https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5 (last access: 25
- 542 <u>May</u> 2021)
- 543

544 Author contributions

- 545 YY designed the research. LZ performed the model simulations and analyzed the data.
- 546 All the authors discussed the results and wrote the paper.
- 547
- 548 *Competing interests*
- 549 The authors declare that they have no conflict of interest.
- 550

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796 **Table 1.** Experimental design.

	Experiments	Model Configuration
	<u>CLIM</u>	Climatological SST
	<u>SD</u>	Climatological SST + Δ SST _{SD El Niño}
	LD	Climatological SST + Δ SST _{LD El Niño}
	SD_emis	Same as SD but turn off the emissions from South and Southeast Asia
	CLIM_emis	Same as CLIM but turn off the emissions from South and Southeast Asia
797		

删除[z]: **Table 1.** The seasonal mean aerosol concentrations (unit: µg m⁻³) and number of haze days (unit: day) in December-January-February (DJF) over various regions of China, including NCP, SCB, YRD, PRD, NEP, YGP, and FWP from CLIM, SD and LD experiments. Haze days are defined as days with daily average near-surface PM_{2.5} concentrations above the 90th percentile in each region. The values in brackets represent the anomalies in SD and LD relative to CLIM.

NEP
YGP
FWP
Mean
Conc.
CLIM
24.87
32.33
27.98
17.26
9.42
20.19
25.11
SD
23.73
(-1.14)
31.16
(-1.17)
28.21
(+0.23)
18.20
(+0.94)
9.95
(+0.53)
19.55
(-0.64)
23.17
(-1.94)
LD
24 76

NCP SCB

YRD PRD





800 Figure 1. Time series of the Niño3.4 index (°C) based on the merged Hadley-NOAA/OI SST

801 dataset for 1870-2017. The time series were detrended and smoothed with a 3-month running

802 average filter. Highlighted slots illustrate the SD (green) and LD (orange) El Niño events.



Figure 2. Time series of the Niño3.4 index (°C) overlaid from Jun⁻¹ to Jun⁺² for (left) SD and 806 (middle) LD El Niño events during 1870-2017. The individual and composite events are shown by 807 808

thin gray and bold red curves, respectively. The total number and percentage of events are shown

at the upper left corner of each panel. A comparison of the composite time series of Niño3.4 index 809 810 for SD and LD events is shown in the right panel.



811 812

813 Figure 3. Composite differences in DJF mean SST (°C) between SD (a) / LD (b) El Niño events

814 and climatological mean over 1870-2017 and between SD and LD (c) El Niño events. Differences

815 that are statistically significant at 95% from a two-tailed T-test are stippled.

816





820 Figure 4. Spatial distributions (a) and scatter plots (b) of observed and simulated DJF mean

821 near-surface $PM_{2.5}$ concentrations ($\mu g m^{-3}$) from the CLIM experiment. Solid line represents 1:1

ratio and dashed lines mark 1:3 and 3:1 ratios. The observed concentrations are derived from the

823 CNEMC in December 2014-February 2015. The normalized mean <u>bias (NMB)</u> and the correlation

删除[z]: deviation

826 827 at the site i, respectively.





833 statistical significance with 90% confidence from a two-tailed T-test.

834

828 829 830

831

删除[z]: Composite

(a) surface conc SD-CLIM

90°E

(d) aerosol burden SD-CLIM

55°N

45°N

35°N

25°N

15°N

55°N

45°N

35°N

25°N

15°N

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75°E

-1.6

75°E

-2.4

 $\mu g/m^3$

-1.2

mg/m²

55°N

-0.8

105°E 120°E 135°E

-1.8

90°E 105°E 120°E 135°E

-1.2

(b) surface co

55°N

45°N

35°N

25°N

15°N

45°N

35°N

25°N

15°N

75°E

-0.4

90°

75°E

-0.6

(e) aerosol bu







Figure 6. Composite differences (%) in DJF mean near-surface PM_{2.5} concentrations and aerosol column burdens between SD and CLIM (a, d), LD and CLIM (b, e), and SD and LD (c, f), relative to CLIM. The stippled areas indicate statistical significance with 90% confidence from a two-tailed T-test.



Figure 6. Composite differences in DJF mean sea level pressure (SLP, shaded; units: hPa) and
wind at 850 hPa (WIND850, vector; units: m s⁻¹) (top panels) and geopotential height at 500 hPa
(GPH500, shaded; units: m) and wind at 500 hPa (WIND500, vector; units: m s⁻¹) (bottom panels)

between SD and CLIM (a, d), LD and CLIM (b, e), and SD and LD (c, f). The stippled areas

842 indicate statistical significance with 90% confidence from a two-tailed T-test.



845 Figure 7. Composite differences in DJF mean winds at 850 hPa (m s⁻¹) (top panels) and 500 hPa

846 (m s⁻¹) (bottom panels) between 2015/2016 SD El Niño and climatological mean (1950-2017) (a,
847 d), 1986/1987 LD El Niño and climatological mean (b, e), and 2015/2016 SD El Niño and

848 1986/1987 LD El Niño (c, f) from the EAR5 reanalysis data. The data were detrended over

- 849 <u>1950-2017.</u>
- 850





854 Figure 8. Composite differences in DJF mean precipitation rate (top panels; units: mm day⁻¹) and wet deposition of PM_{2.5} (bottom panels; units: mg m⁻² d⁻¹) between SD and CLIM (a, d), LD and 855 856 CLIM (b, e), and SD and LD (c, f). The stippled areas indicate statistical significance with 90% confidence from a two-tailed T-test. 857



Figure 9. Composite differences in DJF mean near-surface $PM_{2.5}$ concentration (µg $m^{\text{-}3})$ and aerosol column burden (mg m⁻²) between SD emis and CLIM emis (a, c) SD and CLIM (b, d).

The stippled areas indicate statistical significance with 90% confidence from a two-tailed T-test.





Figure 10. (a) Subregions of China defined in this study, including the North China Plain (NCP, 35-41°N, 114-120°E), the Sichuan Basin (SCB, 28-33°N, 103-108°E), the Yangtze River Delta (YRD, 29-34°N, 118-121.5°E), the Pearl River Delta (PRD, 21.5-25°N, 111-116°E), the Northeast Plain (NEP, 41-48°N, 120-130°E), the Yunnan-Guizhou Plateau (YGP, 23-27°N, 100-110°E), and the Fenwei Plain (FWP, 33-35°N, 106-112°E and 35-38°N, 110-114°E). (b-h) Probability density distributions of daily near-surface $PM_{2.5}$ concentrations (µg m⁻³) in DJF over various subregions of China. The seasonal mean aerosol concentrations (unit: µg m⁻³) and number of haze days (unit: day) in December-January-February (DJF) over various regions of China from CLIM, SD and LD experiments are shown in the corresponding table. Haze days are defined as days with daily average near-surface PM2.5 concentrations above the 90th percentile in each region. The values in brackets represent the anomalies in SD and LD relative to CLIM.



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Figure 11. Stacked histograms of the number of SD and LD El Niño events per 15 years (except 1870-1880 for 10 years) during 1870-2015. The red and blue dashed lines indicate linear trends in the number of SD and LD El Niño events, respectively. Their p-values are shown in the upper right corner of the figure, which indicate the increasing trend of SD at a two-tailed T-test confidence level of 94% for 1870-2015 (87% for 1880-2015 and 92% for 1940-2015) statistical significance.



Figure 11. Probability density distributions of daily near-surface $PM_{2.5}$ concentrations (µg m⁻³) in DJF over various subregions of China.

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