1	A seesaw haze pollution in North China modulated by
2	sub-seasonal variability of atmospheric circulation
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#### Abstract

Utilizing a recent observational particulate matter with diameters less than 2.5 µm 27 28 (PM<sub>2.5</sub>) dataset in North China, this study reveals a distinct seesaw feature of abnormally high and low PM2.5 concentrations in the adjacent two months of 29 December 2015 and January 2016, accompanied by distinct meteorological 30 modulations. The seesaw pattern is postulated to be linked to super El Niño and the 31 Arctic Oscillation (AO). During the mature phase of El Niño in December 2015, the 32 33 weakened East Asian Winter Monsoon (EAWM) and the associated low-level southerly wind anomaly reduced planetary boundary layer height, favoring strong 34 haze formation. This circulation pattern was completely reversed in the following 35 36 month, in part due to a sudden phase change of the AO from positive to negative and the beginning of a decay of the El Niño, which enhanced the southward shift of the 37 upper-tropospheric jet from December to January relative to climatology, leading to 38 an enhanced EAWM and substantially lower haze formation. This sub-seasonal 39 40 change in circulation is also robustly found in 1982/1983 and 1997/1998, implicative of a general physical mechanism dynamically linked to El Niño and the AO. 41 Numerical experiments with the Weather Research and Forecasting 42 (WRF)-Community Multi-scale Air Quality (CMAQ) model were used to test the 43 44 modulation of the meteorological conditions on haze formation. With the same emission, simulations for three super El Niño periods (1983, 1997 and 2015) robustly 45 show higher PM2.5 concentrations under the mature phase of the super El Niño, but 46 47 substantially lower PM<sub>2.5</sub> concentrations during the decay phase of El Niño (and the sudden AO phase change), further verifying the modulation effect of sub-seasonal 48 circulation anomaly on PM2.5 concentrations in North China. 49 Key words: Haze, El Niño, Arctic Oscillation, East Asian winter monsoon, 50

51 WRF/CMAQ

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# 53 1. Introduction

China has experienced severe haze pollution in recent years (Ding et al., 54 2016;Song et al., 2017;Tie et al., 2017;Zhang et al., 2015), with potentially 55 detrimental effect on human health (Chen et al., 2017) as suggested by a substantial 56 increase in hospital visits during the haze season (Liang et al., 2017;Xu et al., 2017c). 57 Understanding the mechanism of haze formation is vital for developing effective 58 measures to relieve the haze pollution. Despite the continued reduction in 59 anthropogenic emission such as NO<sub>x</sub>, SO<sub>2</sub>, and CO in China in the past few years (Li 60 et al., 2017;Liu et al., 2016;Wang et al., 2017), the severe haze pollution motivates a 61 need to understand the mechanism of haze distribution from a meteorological 62 perspective (Ding et al., 2016;He et al., 2017). A few studies investigated the 63 relationship between haze and climate variability at decadal or longer time scales 64 (Jeong and Park, 2017;Liu et al., 2017;Wu et al., 2015;Xu et al., 2017a;Yin et al., 65 2017; Zhang et al., 2016), while others examined the meteorological factors associated 66 67 with specific severe haze events (Cai et al., 2017;Li et al., 2018;Yin et al., 2017).

By using a long-term observational data set from 1972-2014, Li et al. (2016) 68 found that the number of fog-haze days in winter across central and eastern China has 69 a strong relationship with the East Asian winter monsoon (EAWM), with weak 70 71 EAWM favoring the accumulation of haze by weakening the near surface wind speed. This effect was further illustrated by Yang et al. (2016) using a twenty-year long 72 simulation from 1985 to 2005 with the Goddard Earth-Observing System chemical 73 transport model (GEOS-Chem). Zooming in on the severe haze event in 2013, Cai et 74 al. (2017) identified the conducive weather conditions for severe haze including 75 weakened surface northerlies and northward shift of mid-troposphere northwesterlies 76 extending to the north of Beijing, reducing the cold and dry flow to Beijing. 77

The recent severe haze event in December 2015 has been linked to the strong El Niño condition (Chang et al., 2016;Yuan et al., 2017;Zhang et al., 2017). For example, Yuan et al. (2017) investigated the impact of El Niño on the severe haze during

November to December of 2015 and found unfavorable meteorological conditions 81 during this period including a weak East Asian winter monsoon, reduced cold air 82 intrusion, decreased low level wind speed and enhanced stability unfavorable for 83 ventilation of pollutants. Similar to El Niño-Southern Oscillation (ENSO) but from a 84 decadal simulation, Zhao et al. (2016) found that the decadal variations of haze days 85 in central-eastern China during 1959-2012 are tightly associated with the Pacific 86 Decadal Oscillation (PDO), with more haze days occurring during positive PDO 87 phase, primarily resulting from enhancement of the Mongolia High and a stable 88 atmosphere, whereas an opposite effect was observed during the negative phase of 89 PDO. The modulation of El Niño on other air pollutants such as ozone has also been 90 investigated. For example, Xu et al. (2017b) found different effects of ENSO 91 modulation on ozone between the developing and decaying phase of ENSO. During 92 the developing phase in fall, El Niño tends to reduce ozone in the southeastern US 93 compared to La Niña, possibly due to the increased water vapor as an ozone sink and 94 cooler surface air temperature and stagnation. In contrast, during the decaying phase 95 96 in spring, ozone in the western US shows some decreases likely linked to the decreased temperature and enhanced water vapor in that region. 97

While it is highly possible that the severe haze pollution in November to 98 99 December of 2015 is a result of the strong El Niño, the sudden drop in PM2.5 concentrations in January 2016 compared to December 2015 has heretofore largely 100 been ignored. Puzzled by the seesaw pattern of severe haze pollution in December 101 102 2015 and the low PM<sub>2.5</sub> concentrations in January 2016, this study aims to explore the possible mechanism behind this temporal seesaw phenomenon. The paper is 103 organized as follows. In Section 2, we introduce the data and methodology used in the 104 study, followed by the monthly variations of PM<sub>2.5</sub> concentration. In Section 4, 105 mechanisms regarding the haze variations are investigated. 106

# 107 2. Data and methodology

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Meteorological data including zonal and meridional wind at 500 hPa and 850

hPa, geopotential height at 500 hPa, 2-m air temperature and planetary boundary layer
height, are from the European Centre for Medium-Range Weather Forecasts
(ECMWF) Interim Reanalysis Data (ERA-Interim) (Dee et al., 2011), with a spatial
resolution of 1.125° by 1.125°. Six hourly data were downloaded to calculate monthly
and daily mean. A thirty-year period of 1987-2016 is selected as the reference period
when anomaly is calculated. The AO index and Niño 3.4 index are available at the
Climate Prediction Center (CPC) website (http://www.cpc.ncep.noaa.gov/).

The PM<sub>2.5</sub> hourly concentrations during 2014-2017, the only period with available data, were downloaded from http://www.pm25.in for more than 1000 stations and the data were interpolated to the same spatial resolution as the ERA-Interim, i.e., 1.125° by 1.125°. A longer-term dataset of PM2.5 concentrations from 2009-2017 at the U.S. Embassy in Beijing (117°E, 40°N), was downloaded from http://www.stateair.net/web/historical/1/1.html.

A regional meteorological model, Weather Research and Forecasting (WRF) 122 (Skamarock et al., 2008) model, coupled to a chemistry model, Community 123 Multi-scale Air Quality (CMAQ), was used to investigate possible factors modulating 124 haze formation. WRF version 3.8.1 was used in this study, with physics options same 125 as those discussed in Gao et al. (2017), i.e., for planetary boundary layer (PBL), the 126 Mellor-Yamada-Janjic scheme was used (Janjić, 1990;Mellor and Yamada, 1982). The 127 domain covers majority of East Asia (shown later; i.e., Fig. 5b), with spatial resolution 128 of 36 km by 36 km. The pressure of the model top is 50 hPa, with lambert conformal 129 conic projection centered at 34°N, 110°E. A total of 34 layers were used, with the top 130 of the first layer at about 40 meters. The initial and boundary conditions for WRF were 131 provided by the NCEP Climate Forecast System Reanalysis (CFSR) version 2 (Saha et 132 al., 2013), with a spatial resolution of 0.5 deg×0.5 deg. For regional chemistry, the 133 widely used CMAQ model (Byun and Ching, 1999; Byun and Schere, 2006), with the 134 latest version 5.2, was used in this study, with the carbon-bond version 6 (CB06) for the 135 major gas phase chemistry and AERO6 for aerosol module. The removal of particulate 136

matter includes dry and wet deposition. For dry deposition, it is in general expressed by 137 the product of dry deposition velocity and concentration of pollutants. The dry 138 deposition velocity is the inverse of resistance including aerodynamic resistance, 139 molecular motion and surface resistance, and more details were discussed in Pleim and 140 Ran (2011). The wet deposition of pollutants depends on their concentrations in cloud 141 water and the precipitation rate, and the algorithms were based on regional acid 142 deposition model (RADM; (Chang et al., 1987)). The latest version of 143 Meteorology-Chemistry Interface Processor (MCIP 4.3) was used to post-process 144 WRF results and prepare input data for CMAQ (Otte and Pleim, 2010). The initial and 145 boundary chemical conditions were derived from Model for Ozone and Related 146 chemical Tracers, version 4 (MOZART-4). Downscaling from MOZART to CMAQ 147 was developed and discussed in detail in an upcoming paper (Ma et al., in preparation). 148 The emissions of air pollutants in China were estimated by Tsinghua University 149 (Wang et al., 2014;Zhao et al., 2013;Zhao et al., 2017;Zhao et al., 2018) using an 150 "emission factor method". The provincial emissions from area and mobile sources 151 were calculated from the activity data (energy consumption, industrial product yields, 152 solvent use, etc.), technology-based uncontrolled emission factors, and penetrations of 153 control technologies, and subsequently distributed to the model grids according to the 154 spatial distribution of population, GDP, and road networks. A unit-based method is 155 applied to estimate and locate the emissions from large point sources including power 156 plants, iron and steel plants, and cement plants. The emissions from open burning of 157 agricultural residue have been included in the anthropogenic emission inventory 158 developed by Tsinghua University. They were calculated based on crop yields, the 159 ratio of residue to crop, the fraction of biomass burned in the open field, and emission 160 factors (Wang and Zhang, 2008; Zhao et al., 2013; Zhao et al., 2018). The emissions 161 from natural forest and grassland fires was ignored in this study, primarily due to the 162 relatively small contribution in particular over the North China (Qin and Xie, 2011). 163 Biogenic emissions were calculated by the Model of Emissions of Gases and Aerosols 164 from Nature (MEGAN; Guenther et al., 2006). For each month of the CMAQ 165

simulations, a week of model spin up was used to reduce the influence of the initialconditions.

# 168 **3. Monthly variations of PM<sub>2.5</sub> concentration**

## 169 3.1 Difference of PM<sub>2.5</sub> in December 2015 and January 2016

Fig. 1 shows the spatial distribution of PM<sub>2.5</sub> concentration in December 2015 (Fig. 170 171 1a) and the monthly mean difference between the anomaly of January 2016 (relative to January from 2015-2017) and December 2015 (relative to December from 172 2014-2016). The three-year average PM<sub>2.5</sub> concentration in December (2014-2016) 173 and January (2015-2017) was shown in Fig. S1 in the supporting information. 174 Consistent with previous studies (Chang et al., 2016; Yuan et al., 2017; Zhang et al., 175 2017), severe haze pollution occurred in December 2015 particularly over the North 176 177 China Plain (NCP) (black box in Fig. 1a excluding Bohai area), with monthly mean PM<sub>2.5</sub> as high as 148  $\mu$ g/m<sup>3</sup>, much higher than that in December 2014 (97  $\mu$ g/m<sup>3</sup>) and 178 slightly higher than that in December 2016 (135 µg/m<sup>3</sup>). In January 2016, the 179 anomaly (relative to January from 2015-2017) of PM2.5 showed large decrease 180 compared to the anomaly of December 2015 (relative to December from 2014-2016) 181 over the NCP, with a mean decrease of 35  $\mu$ g/m<sup>3</sup>, whereas the PM<sub>2.5</sub> anomalies 182 increase in January of 2015 (33  $\mu$ g/m<sup>3</sup>) and 2017 (3  $\mu$ g/m<sup>3</sup>) relative to the adjacent 183 December anomaly (Fig. S2 in the supporting information). 184

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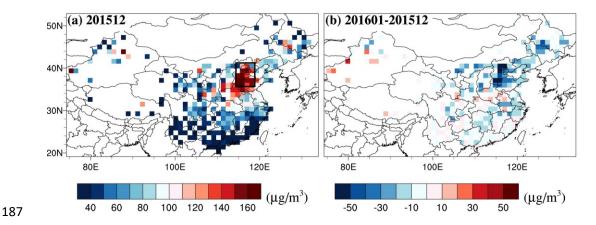


Fig. 1. Monthly mean PM<sub>2.5</sub> concentration in December 2015 (Fig. 1a), and the difference (Fig. 1b)
between the anomaly of January 2016 (relative to January from 2015-2017) and December 2015
(relative to December from 2014-2016). The black box in Fig. 1a indicates the area of NCP.

## 3.2 PM<sub>2.5</sub> anomaly from long time series

To further test the robustness of the large drop of PM2.5 from December 2015 to 192 January 2016, monthly mean anomaly of PM2.5 relative to 2009-2017 over U.S. 193 Embassy Beijing is shown in Fig. 2. The PM<sub>2.5</sub> anomaly in December is generally 194 negative (i.e., low PM<sub>2.5</sub> concentration) from 2009-2014 except for a substantial 195 positive value of 48  $\mu$ g/m<sup>3</sup> in 2015. For the January PM<sub>2.5</sub> anomaly, the large positive 196 value in 2013 has been widely studied to investigate the mechanisms modulating the 197 severe haze events (Han et al., 2016;Kajino et al., 2017;Shi et al., 2017;Ye et al., 198 2016). Looking at the difference between the January and December anomalies, the 199 exceptional difference between January 2016 and December 2015 stands out, with the 200 PM<sub>2.5</sub> concentration anomaly (-38  $\mu$ g/m<sup>3</sup>) in January 2016 showing a decrease of -86 201  $\mu g/m^3$  relative to the anomaly in December 2015 (48  $\mu g/m^3$ ), which is much more 202 negative than the difference in any other years and far more negative than the 99% 203 204 confidence interval value of -66  $\mu$ g/m<sup>3</sup>.

In addition to the single site of U.S. Embassy Beijing, two other data sets of air pollution index (API; http://datacenter.mep.gov.cn/) and air quality index (AQI; http://www.pm25.in) were combined to illustrate the robustness of the abnormal difference between January and December PM2.5 over a larger spatial area in NCP

(black box in Fig. 1). Combining the API and AOI data allows anomaly to be 209 calculated relative to a longer period despite the short record of the AQI data. Similar 210 to Fig. 2, abnormally high concentration in December 2015 and low concentration in 211 January 2016 are also found in the API/AQI record (Fig. S3 in the supporting 212 information). As anthropogenic emissions such as NO<sub>x</sub>, SO<sub>2</sub>, and primary PM<sub>2.5</sub> 213 have been steadily decreasing since 2011 (Liu et al., 2016; Wang et al., 2017), the 214 abnormally high PM<sub>2.5</sub> concentration in December 2015 requires an explanation. 215 Moreover, the anthropogenic emissions in January could be comparable to or higher 216 than that in December, i.e., in January 2016, higher SO<sub>2</sub> concentration, implicative of 217 SO<sub>2</sub> emissions, was found than December 2015 based on observed data 218 (http://www.pm25.in; not shown). Thus, what mechanism triggers the sharp decrease 219 of haze in January 2016 compared to December 2015 needs to be investigated. 220

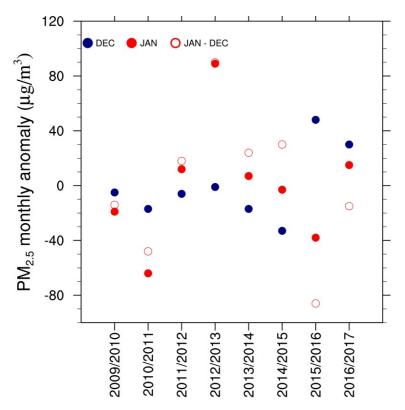


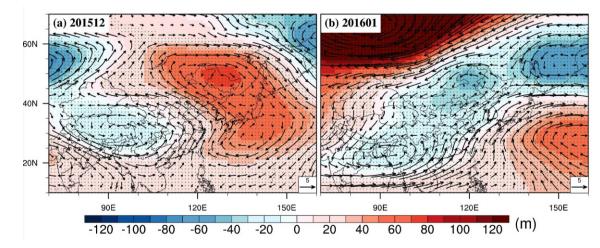
Fig. 2. Monthly mean anomaly of PM<sub>2.5</sub> in the adjacent December (solid blue) and
January (solid red) as well as the change (hollow red circle) from December to
January during 2009-2017 over US embassy Beijing. For December (January), the
anomaly is relative to 2009-2016 (2010-2017) eight-year respective monthly mean

#### value.

# 4. Mechanism modulating the high and low anomaly of PM<sub>2.5</sub>

# 227 4.1 The effect of meteorological modulation

To determine if meteorological factors play a role in the anomalous December to 228 January change in PM<sub>2.5</sub>, we first examined the mid-tropospheric circulation system 229 during December 2015 and January 2016. As shown in Fig. 3a for December 2015, 230 the northeastward shift of Siberian High and anticyclonic high pressure system in the 231 NCP reduced the northerly wind transporting cold air from Siberia to the NCP, 232 favoring the haze formation (Cai et al., 2017; Chang et al., 2016). In January 2016 (Fig. 233 3b), a low pressure system dominates over the NCP area, enhancing the northerly 234 wind blowing from Siberia and relieving haze formation. An important question here 235 236 is whether the transition of the meteorological conditions from December 2015 to January 2016 was influenced by climate variations on seasonal-to-decadal time scales 237 with some level of predictability. 238



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Fig. 3 Anomaly of geopotential height and wind vector at 500 hPa (unit: m/s) for December 2015 (Fig. 3a) and January 2016 (Fig. 3b), with anomaly relative to the respective month in1987-2016. Stippled areas indicate geopotential height exceeding 90<sup>th</sup> confidence interval. The 90th confidence interval was calculated for each grid based on a two tailed t-distribution method using the respective month data over the

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thirty-year (1987-2016) period (i.e., geopotential height or wind vector at 500 hPa). The black box in Fig. 3a indicates the area of NCP.

#### 247 **4.2 Possible linkage with climate variability**

From the Nino 3.4 index, defined as the 3-month running regional mean SST 248 anomaly over the tropical region (5 °S-5 °N, 170°-120°W) relative to 1951-2000 249 (Rayner et al., 2003), the El Niño signal reached a peak in December 2015, and 250 started to decay in January 2016. The development and decay of this El Niño event 251 and the associated change in circulation pattern have been well documented by Xue 252 and Kumar (2017). The 2015-2016 El Niño event is known as a super strong event 253 comparable to the other two super events during 1982/1983 and 1997/1998 (Ren et al., 254 255 2017). Thus, it is appropriate to ask whether the circulation pattern in the NCP was modulated by the development of a super El Niño. Motivated by this question, we 256 examined the super El Niño events in 1982/1983 and 1997/1998 based on Nino 3.4. In 257 1997/1998, the El Niño peaked in December 1997 and started to decay in January 258 1998, but in 1982/1983, the El Niño peaked in January 1983 and started to decay in 259 February 1983. The 500 hPa geopotential height anomalies during these four months 260 are shown in Fig. S4. Consistent with the anomalous circulation features in Fig. 3, an 261 anticyclonic circulation anomaly dominates over the NCP during the peak of the El 262 263 Niño (199712, 198301) while a cyclonic circulation anomaly prevails over the same region when the El Niño started to decay (199801, 198302). To exclude this opposite 264 anomaly patterns as climatological features, we checked the 500 hPa anomalies in 265 December, January, and February during the past thirty years (1987-2016) but we 266 could not find similar opposite patterns in adjacent winter months during any other 267 years. Moreover, the ensemble mean circulation anomaly during the peak and decay 268 of the El Niño events from 1948-2016 was shown in Fig. S5 in the supporting 269 information, and only the composite of three super El Nino events 270 271 (1982/1983,1997/1998, 2015/2016; Figs. S5a,b) shows the seesaw patterns, whereas the other ensemble results did not show such unique feature (Figs. S5c-f). This further 272

hints at the possible linkage between the anomalous circulation patterns and super ElNiño.

275 Since low-level wind has a larger influence on the formation of haze than mid-tropospheric wind, the composited anomaly of 850 hPa wind vector and near 276 surface air temperature during the adjacent months of the super El Niño events 277 (January 1983, December 1997 and 2015 versus February 1983 and January 1998 and 278 2016) are shown in Fig. 4, while anomaly of near surface wind at 10-m is shown in 279 Fig. S6 in the supporting information. These figures clearly depict opposite anomaly 280 patterns in the NCP, showing southerly anomaly (Fig. 4a; Fig. S6a) during the peak of 281 the El Niño, and northerly anomaly (Fig. 4b, Fig. S6b) during the start of the decay 282 283 phase. The southerly wind anomaly, abnormally warm near surface air temperature and stagnant weather conditions over the NCP are indicative of weakened EAWM 284 (Hui and Xiang, 2015) partly related to the warmer air temperature over the northern 285 plain and Siberia and reduced pressure contrast between the Asian continent and the 286 287 western Pacific Ocean, favoring haze formation (Cai et al., 2017;Li et al., 2018). In contrast, there is enhanced northerly flow and more cold air advection (Fig. 4b, Fig. 288 S6b) when El Niño starts to decay. The low-level circulation patterns and near surface 289 air temperature are consistent with the seesaw changes of 500 hPa geopotential height 290 291 from the peak of the El Niño development to the beginning of its decay for all three super El Niño events, highly implicative of a strong relationship between El Niño and 292 haze formation. 293

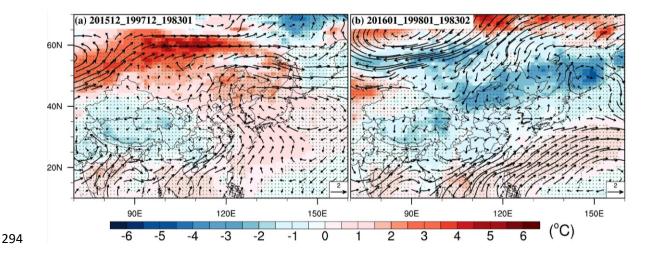


Fig. 4 850 hPa wind anomaly (unit: m/s) and 2-m air temperature anomaly (relative to
1987-2016). The anomaly was conducted over the respective month from 1987-2016,
i.e., mean value of anomaly over December 2015, December 1997 and January 1983
(Fig. 4a), and January 2016, January 1998 and February 1983 (Fig. 4b). Stippled areas
indicate 2-m air temperature exceeding 90<sup>th</sup> confidence interval (the same method as
Fig. 3).

## **4.3 Sensitivities of PM<sub>2.5</sub> to meteorological conditions**

As shown in the above analyses, the meteorological conditions in December 2015, 302 December 1997, January 1983 exhibit a clear contrast with those of the following 303 304 month. To demonstrate a connection between the meteorological conditions and haze formation, we used the WRF/CMAQ regional model to simulate haze for the three 305 time periods under the same emissions, with a spatial resolution of 36 km by 36 km. 306 The simulated meteorological conditions including near surface (2-meter) air 307 temperature (T2) and specific humidity (Q2), 10-m wind speed (WS10) and direction 308 (WD10) were evaluated using the NCEP Meteorological Assimilation Data Ingest 309 System (MADIS; https://madis.noaa.gov) data on hourly time scale. Using the 310 benchmark based on Emery et al. (2001), the meteorological parameters compared 311 312 reasonably well with observations and mostly fall within or quite close to the benchmark, shown in the Table 1 below. Please note that the slightly larger bias 313 (19.25°) of wind direction at 10-m (WD10) in December 2015 is partly attributable to 314

the model comparison with observed values close to  $0^{\circ}$  or  $360^{\circ}$ , which may yield large bias albeit the small differences in reality (i.e.,  $10^{\circ}$  in model vs.  $350^{\circ}$  in observation).

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Table 1 Evaluation of WRF performance over North China (9 stations)<sup>\*</sup>

NCP	201512				201601			Benchmark (Emery et al., 2001)				
	T2	Q2	WD10	WS10	T2	Q2	WD10	WS10	T2	Q2	WD10	WS10
Bias	0.31	0.24	19.25	0.96	0.07	0.25	1.48	0.92	<±0.5	< ±1	<±10	<±0.5
Gross	2.31	0.63	88.36	1.57	2.30	0.54	85.39	1.79	< 2	< 2	< 30	/
Error												
RMSE	2.99	0.92	135.66	2.09	2.97	0.80	137.09	2.30	/	/	/	< 2

\* T2: temperature at 2-mter with unit of °C

320 Q2: specific humidity with unit of g/kg

WD10: wind direction at 10-meter with unit of degrees

WS10: wind speed at 10-meter with unit of m/s

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MCIP was used to prepare input for CMAQ. A code bug in MCIP 4.3 for 324 processing land use information was found that affected the percentage of urban area 325 (PURB), leading to low PM2.5 concentration in the simulations over urban areas (see 326 the part 2 of the supporting information for more details). With the bug fix, the 327 concentration of PM2.5 matches the observations well (Fig. 5). Statistical metrics such 328 as mean fractional bias/error (MFB/MFE) were used to evaluate the simulation of 329 PM<sub>2.5</sub>, as recommended by US EPA (USEPA, 2007). Based on almost 200 330 observational sites in North China, the MFB/MFE of PM2.5 from CMAQ is 1%/55% 331 and 1%/56%, respectively for December 2015 and January 2016, satisfying the 332 benchmark of 50%/75% (USEPA, 2007). More evaluation of PM2.5 was discussed in 333 part 3 of the supporting information. 334

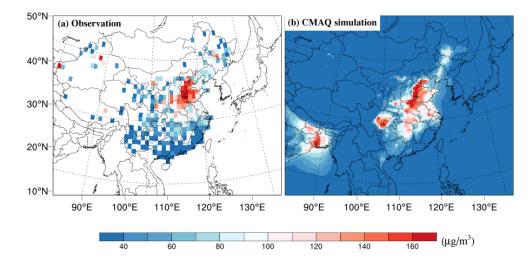
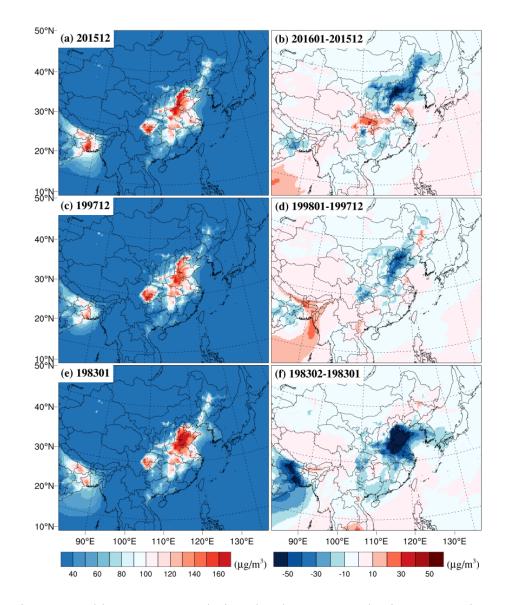






Fig. 5. Spatial distributions of monthly mean PM2.5 concentration in December 2015 for observations (a) and CMAQ simulation (b). Please note that the label bar starts from 40  $\mu$ g/m<sup>3</sup> instead of 0 in order to make the comparison easily viewed. 339

Fig. 6 shows the spatial distribution of monthly mean PM<sub>2.5</sub> in December 2015 340 (Fig. 6a), December 1997 (Fig. 6c), January 1983 (Fig. 6e) and the differences 341 compared to the PM<sub>2.5</sub> in the following month. Although the same emissions at the 342 level of December 2015 were used across the simulations, the results clearly show 343 much higher monthly mean PM<sub>2.5</sub> in December 2015 (Fig. 6a), December 1997 (Fig. 344 6c), January 1983 (Fig. 6e) particularly over the NCP compared to the following 345 month (Figs. 6b,d,f), with mean reduction in the NCP of 30-50  $\mu$ g/m<sup>3</sup> or more. In 346 addition, by applying the process analysis (not shown), we found that the dry and wet 347 deposition contributed a total of 12 % to the total removal processes over NCP in 348 December 2015, comparable to that (13%) in January 2013, indicating the seesaw 349 patterns was not modulated by the deposition. Hence the modeling results further 350 verified the effect of the meteorological conditions on the seesaw PM<sub>2.5</sub> pattern. 351



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Fig. 6 Monthly mean PM<sub>2.5</sub> during the three super El Niño events, i.e., December 2015 (a), December 1997 (c), January 1983 (e) and the differences of PM<sub>2.5</sub> compared to the following month (b, d, and f)

# 357 4.4 Synthesis of the mechanism modulating the PM<sub>2.5</sub> variations

358 The different modulation effects of the development and decay phase of El Niño

- 359 on East Asian summer monsoon circulation have been noted in previous studies (Xue
- et al., 2018; Yuan and Yang, 2012). For example, Xue et al. (2018) discussed a
- 361 mechanism for how developing El Niño in the summer modulates the intraseasonal

variation of the West Pacific Subtropical High (WPSH). They found that compared to 362 June and July, enhanced convection in August over the warm pool significantly 363 reduces the 500 hPa geopotential height and pushes the WPSH to retreat eastward 364 substantially. For intraseasonal variation in winter, Nie et al. (2016) found that the 365 change from warm anomaly in December 2015 to cold anomaly in January 2016 in 366 the NCP is possibly associated with the sudden shift of the Arctic Oscillation (AO) 367 from a positive phase to a negative phase in January 2016, which is dynamically 368 linked to super El Niño (Geng et al., 2017). The impact of the AO on the EAWM was 369 also demonstrated in December 2012 when the Siberian High and cold advection were 370 strong during the negative phase of the AO, but a transition of the AO to a positive 371 phase in January 2013 caused a sudden weakening of cold advection and increased 372 stagnation. The resulting severe haze in January 2013 has been widely discussed 373 previously (Cheng et al., 2017;Kajino et al., 2017). 374

The EAWM is closely related to winter haze conditions in the NCP. Cheung et al. 375 376 (2012) found that in general, the EAWM is weakened (enhanced) when the AO and ENSO are in phase (out of phase). They also noted that a similar relationship at 377 sub-seasonal time scale may be further investigated considering the possibility of a 378 sharp reversal of the AO resulting from tropospheric-stratospheric interaction 379 (Baldwin and Dunkerton, 1999). To delve into the mechanism modulating the sudden 380 reversal of the AO from positive to negative phase during the decay of the mature 381 phase of El Niño, anomaly zonal wind changes at 200 hPa are shown in Fig. 7. 382 Climatologically (i.e., 1987-2016), the subtropical jet is centered around 35-40°N in 383 December and shift southward in January based on ERA-Interim (not shown). A clear 384 dipole feature of the sub-seasonal changes of anomaly zonal wind with negative 385 change north of 40°N indicates an obvious anomalous sub-seasonal southward shift of 386 the subtropical jet during the three El Niño events from its climatological 387 sub-seasonal shift. The subtropical jet shift has been confirmed by Geng et al. (2017) 388 using climate model simulations, showing phase transition of AO from positive to 389

negative in early January during super El Niño winter. The southward shift of the 390 upper tropospheric jet in January 2016 is likely associated with the weakened 391 stratospheric polar vortex and the subsequent negative phase of the AO (Bell et al., 392 2009;Fletcher and Kushner, 2010), leading to more cold advection, enhanced EAWM, 393 and higher PBL height compared to the conditions during the peak of the El Niño with 394 lower PBL height (Fig. 8a). These changes in the meteorological conditions 395 associated with the changes in the El Niño development and AO phase shift and the 396 397 consequent changes in PM2.5 concentration are summarized schematically in Fig. 9. In short, during the mature phase of El Niño (Fig 9a; indicated by the red Nino 3.4 index 398 in the bottom of Fig. 9), the El Niño and positive AO (+AO) are in phase and weakens 399 the EAWM (with orange and blue arrows indicating effect from El Nino and AO, 400 respectively in Fig. 9a), leading to lower PBL and the subsequent severe haze 401 pollution. After the mature phase of El Niño (Fig. 9b), AO suddenly turns to the 402 negative phase (-AO), enhancing the EAWM (blue arrows in Fig. 9b) and subduing 403 the weakening effect from El Nino (the southerly orange arrow) on EAWM, resulting 404 405 in low PM<sub>2.5</sub> concentration.

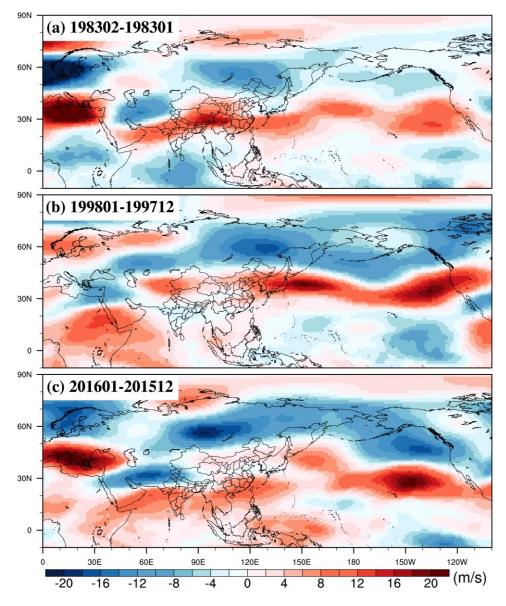


Fig. 7 Sub-seasonal changes of anomaly of 200 hPa zonal wind (relative to the
respective month in 1987-2016) during the start of the decay for three super El Niño
events, including the differences between anomaly of February 1983 and January
1983 (Fig. 7a), January 1998 and December 1997 (Fig. 7b), January 2016 and
December 2015 (Fig. 7c)

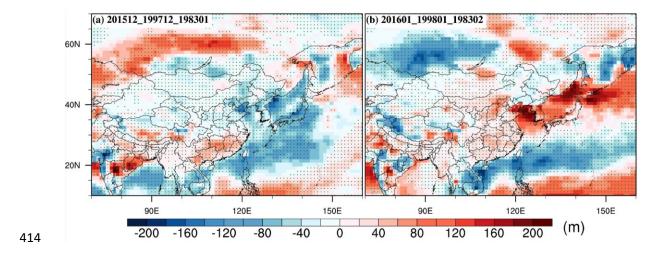


Fig. 8 Sub-seasonal changes of PBL height anomaly (relative to 1987-2016). The
anomaly was conducted over the respective month from 1987-2016, i.e., mean value
of anomaly over December 2015, December 1997 and January 1983 (Fig. 8a), and
January 2016, January 1998 and February 1983 (Fig. 8b). Stippled areas indicate
exceedance of 90<sup>th</sup> confidence interval, with the same method used as Fig. 3.

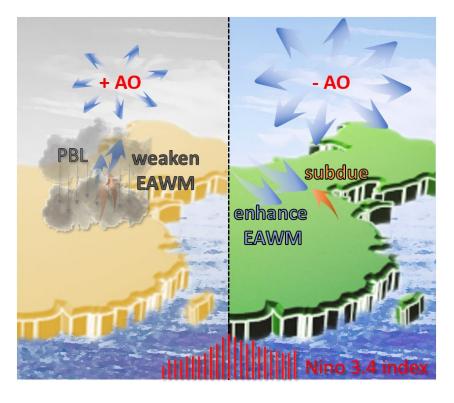


Fig. 9 Schematic of the modulation of El Niño and Arctic Oscillation on

422 atmospheric circulation and haze in northern China

# 424 Summary and discussions

425	This study identifies a distinct seesaw feature of abnormally high and low monthly
426	mean $PM_{2.5}$ concentration in the two consecutive months of December 2015 and
427	January 2016, respectively, in the North China Plain. Accompanying the seesaw
428	features of PM <sub>2.5</sub> are opposite large scale circulation patterns with positive (negative)
429	anomaly of 500 hPa geopotential height and southerly (northerly) wind anomalies at
430	the low level (850 hPa) over the North China Plain during December 2015 (January
431	2016). The contrast in PM <sub>2.5</sub> between December 2015 and January 2016 is
432	significantly larger than the change from December to January between 2009 and
433	2017 recorded at US embassy Beijing.
434	As the modulation of meteorological conditions on PM2.5 occurred during a strong
435	El Niño period, we explored the relationship between strong El Niño and PM2.5. First,
436	using a regional climate/chemistry model WRF/CMAQ, we identified that all three
437	super El Niño events in the recent record (1982/83, 1997/98, and 2015/16) yield
438	similar seesaw modulation features of PM2.5. Further analysis showed that the seesaw
439	PM2.5 variations are modulated by the combined effect of El Niño and Arctic
440	Oscillation (AO). In December 2015, the mature phase of an extreme El Niño,
441	accompanied by a positive AO, weakened the EAWM, as indicated by a positive
442	anomaly of geopotential height at mid-troposphere (i.e., 500 hPa) and southerly wind
443	anomalies at the low level (i.e., 850 hPa) over the North China Plain (NCP), resulting
444	in reduced planetary boundary layer (PBL) height, abnormally warm temperature and
445	substantial haze accumulation during this period. In the following month (January
446	2016) when the El Niño began to decay, a sharp reversal of the AO from a positive
447	phase (in December 2015) to a negative phase triggered enhanced EAWM, inducing
448	cold advection and anomalous low-level northerly winds over the NCP that fostered
449	atmospheric dispersion and substantially reduced haze formation. This abrupt change
450	of the AO from a positive to a negative phase was robustly found for the other two

extreme El Niño events during the corresponding decay period (February 1983 and 451 January 1998) after the mature phase (January 1983 and December 1997). These 452 changes in circulation are likely associated with a southward shift of the upper 453 troposphere jet at 200 hPa and weakened stratospheric vortex during the decay phase 454 compared to the mature phase of El Niño. As the frequency of super El Niño like the 455 2015/16 event is projected to increase in the future (Cai et al., 2014;Cai et al., 2015), 456 the seesaw modulation of super El Niño and AO may become more frequent, 457 458 revealing vital information useful for policy makers dealing with air quality issues in 459 China.

Although we performed WRF-CMAQ simulations to demonstrate the impacts of 460 461 atmospheric circulation during three super El Niño events on haze, this study did not isolate the general effect of El Niño and AO on haze formation. To address this 462 limitation, future studies will design Atmospheric Model Intercomparison Project 463 (AMIP) type scenarios, e.g., by running multi-ensemble members of scenarios with 464 465 global models such as Community Earth System Model (CESM) using different SSTs 466 over the Nino 3.4 area combined with dynamical downscaling using regional climate/chemistry models such as WRF/CMAQ to elucidate the impact of El Niño 467 and AO on haze formation. 468

469 **Competing interests.** The authors declare that they have no conflict of interest.

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