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 PM<sub>2.5</sub> concentrations in the adjacent two months of 29 30 December 2015 and January 2016, accompanied by distinct meteorological modulations. The seesaw pattern is postulated to be linked to super El Niño and the 31 32 Arctic Oscillation (AO). During the mature phase of El Niño in December 2015, the 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  $PM_{2.5}$  concentrations during the decay phase of El Niño (and the sudden AO phase change), further verifying the modulation effect of sub-seasonal 48 49 circulation anomaly on PM<sub>2.5</sub> concentrations in North China. 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 88 phase, primarily resulting from enhancement of the Mongolia High and a stable 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 PM<sub>2.5</sub> 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 2015 and the low PM<sub>2.5</sub> concentrations in January 2016, this study aims to explore the 102 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 3. In 105 Section 4, 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 151 152 were calculated from the activity data (energy consumption, industrial product yields, 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 159 160 ratio of residue to crop, the fraction of biomass burned in the open field, and emission 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 1a) and the monthly mean difference between the anomaly of January 2016 (relative 171 to January from 2015-2017) and December 2015 (relative to December from 172 2014-2016). The three-year average  $PM_{2.5}$  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 China Plain (NCP) (black box in Fig. 1a excluding Bohai area), with monthly mean 177 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 179 180 anomaly (relative to January from 2015-2017) of PM2.5 showed large decrease 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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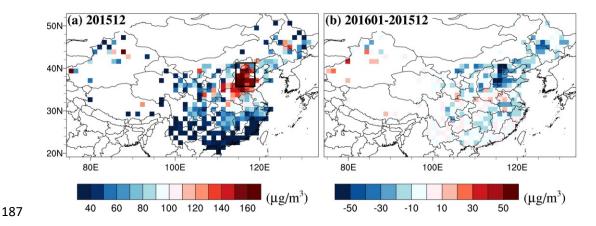


Fig. 1. Monthly mean PM<sub>2.5</sub> concentration in December 2015 (Fig. 1a), and the difference (Fig. 1b)
between the anomaly of January 2016 (relative to January from 2015-2017) and December 2015
(relative to December from 2014-2016). The black box in Fig. 1a indicates the area of NCP.

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

To further test the robustness of the large drop of PM<sub>2.5</sub> from December 2015 to 192 January 2016, monthly mean anomaly of PM<sub>2.5</sub> 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 199 200 exceptional difference between January 2016 and December 2015 stands out, with the  $PM_{2.5}$  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 AQI 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. 215 216 Moreover, the anthropogenic emissions in January could be comparable to or higher than that in December, i.e., in January 2016, higher SO<sub>2</sub> concentration, implicative of 217 was found than December 2015 based on observed data 218  $SO_2$  emissions, (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

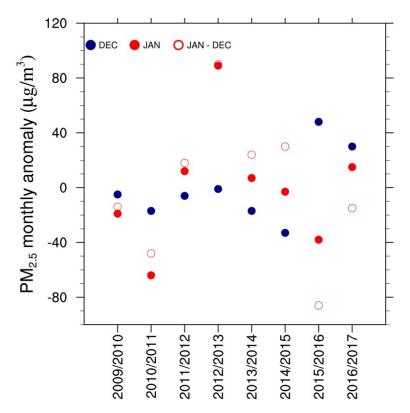


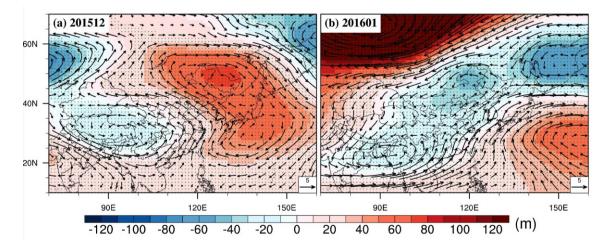
Fig. 2. Monthly mean anomaly of PM<sub>2.5</sub> in the adjacent December (solid blue) and
January (solid red) as well as the change (hollow red circle) from December to
January during 2009-2017 over US embassy Beijing. For December (January), the
anomaly is relative to 2009-2016 (2010-2017) eight-year 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. 233 234 3b), a low pressure system dominates over the NCP area, enhancing the northerly 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

thirty-year (1987-2016) period (i.e., geopotential height or wind vector at 500 hPa).

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# 4.2 Possible linkage with climate variability

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

273 Niño.

Since low-level wind has a larger influence on the formation of haze than 274 275 mid-tropospheric wind, the composited anomaly of 850 hPa wind vector and near surface air temperature during the adjacent months of the super El Niño events 276 (January 1983, December 1997 and 2015 versus February 1983 and January 1998 and 277 2016) are shown in Fig. 4, while anomaly of near surface wind at 10-m is shown in 278 Fig. S6 in the supporting information. These figures clearly depict opposite anomaly 279 280 patterns in the NCP, showing southerly anomaly (Fig. 4a; Fig. S6a) during the peak of the El Niño, and northerly anomaly (Fig. 4b, Fig. S6b) during the start of the decay 281 phase. The southerly wind anomaly, abnormally warm near surface air temperature 282 283 and stagnant weather conditions over the NCP are indicative of weakened EAWM (Hui and Xiang, 2015) partly related to the warmer air temperature over the northern 284 plain and Siberia and reduced pressure contrast between the Asian continent and the 285 western Pacific Ocean, favoring haze formation (Cai et al., 2017;Li et al., 2018). In 286 287 contrast, there is enhanced northerly flow and more cold air advection (Fig. 4b, Fig. S6b) when El Niño starts to decay. The low-level circulation patterns and near surface 288 air temperature are consistent with the seesaw changes of 500 hPa geopotential height 289 from the peak of the El Niño development to the beginning of its decay for all three 290 291 super El Niño events, highly implicative of a strong relationship between El Niño and haze formation. 292

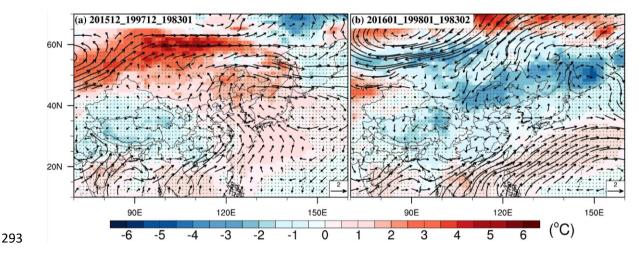


Fig. 4 850 hPa wind anomaly (unit: m/s) and 2-m air temperature anomaly (relative to 1987-2016). The anomaly was conducted over the respective month from 1987-2016, i.e., mean value of anomaly over December 2015, December 1997 and January 1983 (Fig. 4a), and January 2016, January 1998 and February 1983 (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 PM2.5 to meteorological conditions**

As shown in the above analyses, the meteorological conditions in December 2015, 301 December 1997, January 1983 exhibit a clear contrast with those of the following 302 month. To demonstrate a connection between the meteorological conditions and haze 303 formation, we used the WRF/CMAQ regional model to simulate haze for the three 304 time periods under the same emissions, with a spatial resolution of 36 km by 36 km. 305 The simulated meteorological conditions including near surface (2-meter) air 306 temperature (T2) and specific humidity (Q2), 10-m wind speed (WS10) and direction 307 (WD10) were evaluated using the NCEP Meteorological Assimilation Data Ingest 308 System (MADIS; https://madis.noaa.gov) data on hourly time scale. Using the 309 benchmark based on Emery et al. (2001), the meteorological parameters compared 310 reasonably well with observations and mostly fall within or quite close to the 311 benchmark, shown in the Table 1 below. Please note that the slightly larger bias 312 (19.25°) of wind direction at 10-m (WD10) in December 2015 is partly attributable to 313 the model comparison with observed values close to 0° or 360°, which may yield 314 large bias albeit the small differences in reality (i.e., 10° in model vs. 350° in 315 observation). 316

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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 Error	2.31	0.63	88.36	1.57	2.30	0.54	85.39	1.79	< 2	< 2	< 30	/
RMSE	2.99	0.92	135.66	2.09	2.97	0.80	137.09	2.30	/	/	/	< 2

Table 1 Evaluation of WRF performance over North China (9 stations)\*

\* T2: temperature at 2-mter with unit of °C
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 327 processing land use information was found that affected the percentage of urban area 328 (PURB), leading to low PM<sub>2.5</sub> concentration in the simulations over urban areas (see 329 the part 2 of the supporting information for more details). With the bug fix, the 330 concentration of PM<sub>2.5</sub> matches the observations well (Fig. 5). Statistical metrics such 331 332 as mean fractional bias/error (MFB/MFE) were used to evaluate the simulation of PM<sub>2.5</sub>, as recommended by US EPA (USEPA, 2007). Based on almost 200 333 observational sites in North China, the MFB/MFE of PM2.5 from CMAQ is 1%/55% 334 and 1%/56%, respectively for December 2015 and January 2016, satisfying the 335 benchmark of 50%/75% (USEPA, 2007). More evaluation of PM2.5 was discussed in 336 part 3 of the supporting information. 337

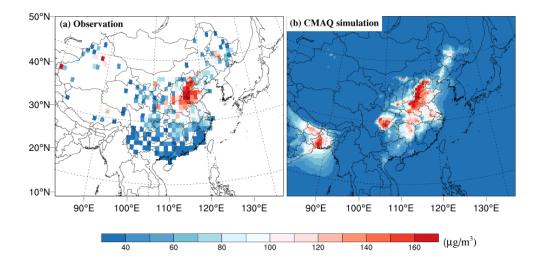
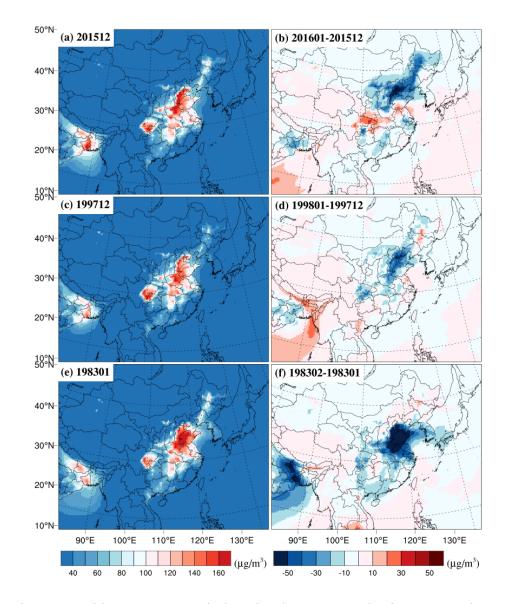






Fig. 5. Spatial distributions of monthly mean  $PM_{2.5}$  concentration in December 2015 for observations (a) and CMAQ simulation (b). Please note that the label bar starts from 40  $\mu$ g/m<sup>3</sup> instead of 0 in order to make the comparison easily viewed.

Fig. 6 shows the spatial distribution of monthly mean PM<sub>2.5</sub> in December 2015 343 (Fig. 6a), December 1997 (Fig. 6c), January 1983 (Fig. 6e) and the differences 344 compared to the PM<sub>2.5</sub> in the following month. Although the same emissions at the 345 level of December 2015 were used across the simulations, the results clearly show 346 much higher monthly mean PM<sub>2.5</sub> in December 2015 (Fig. 6a), December 1997 (Fig. 347 6c), January 1983 (Fig. 6e) particularly over the NCP compared to the following 348 month (Figs. 6b,d,f), with mean reduction in the NCP of 30-50  $\mu$ g/m<sup>3</sup> or more. In 349 addition, we applied the process analysis (Kwok et al., 2013), including horizontal 350 and vertical transport, gas phase chemistry, aerosol process, cloud process, dry 351 deposition and emission to investigate the effect of different processes on PM2.5. The 352 dominant PM2.5 enhancement processes in both December 2015 and January 2016 are 353 emission and aerosol processes. Regarding the removal process, the dominant process 354 355 is the vertical transport, followed by the dry and wet deposition (inferred from cloud process). Both the dry and wet deposition contributed a total of 12% to the total 356 removal processes over NCP in December 2015, comparable to that (13%) in January 357 2016, indicating the seesaw pattern was not modulated by the deposition. Hence the 358 modeling results further verified the effect of the meteorological conditions on the 359 seesaw PM<sub>2.5</sub> pattern. 360



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Fig. 6 Monthly mean  $PM_{2.5}$  during the three super El Niño events, i.e., December 2015 (a), December 1997 (c), January 1983 (e) and the differences of  $PM_{2.5}$  compared to the following month (b, d, and f)

## **366 4.4 Synthesis of the mechanism modulating the PM<sub>2.5</sub> variations**

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

- 368 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
- 370 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 371 June and July, enhanced convection in August over the warm pool significantly 372 reduces the 500 hPa geopotential height and pushes the WPSH to retreat eastward 373 substantially. For intraseasonal variation in winter, Nie et al. (2016) found that the 374 change from warm anomaly in December 2015 to cold anomaly in January 2016 in 375 the NCP is possibly associated with the sudden shift of the Arctic Oscillation (AO) 376 from a positive phase to a negative phase in January 2016, which is dynamically 377 378 linked to super El Niño (Geng et al., 2017). The impact of the AO on the EAWM was also demonstrated in December 2012 when the Siberian High and cold advection were 379 strong during the negative phase of the AO, but a transition of the AO to a positive 380 phase in January 2013 caused a sudden weakening of cold advection and increased 381 stagnation. The resulting severe haze in January 2013 has been widely discussed 382 previously (Cheng et al., 2017;Kajino et al., 2017). 383

The EAWM is closely related to winter haze conditions in the NCP. Cheung et al. 384 385 (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 386 sub-seasonal time scale may be further investigated considering the possibility of a 387 sharp reversal of the AO resulting from tropospheric-stratospheric interaction 388 (Baldwin and Dunkerton, 1999). To delve into the mechanism modulating the sudden 389 reversal of the AO from positive to negative phase during the decay of the mature 390 phase of El Niño, anomaly zonal wind changes at 200 hPa are shown in Fig. 7. 391 Climatologically (i.e., 1987-2016), the subtropical jet is centered around 35-40°N in 392 December and shifts southward in January based on ERA-Interim (not shown). A 393 clear dipole feature of the sub-seasonal changes of anomaly zonal wind with negative 394 change north of 40°N indicates an obvious anomalous sub-seasonal southward shift of 395 the subtropical jet during the three El Niño events from its climatological 396 sub-seasonal shift. The subtropical jet shift has been confirmed by Geng et al. (2017) 397 using climate model simulations, showing phase transition of AO from positive to 398

399 negative in early January during super El Niño winter. The southward shift of the upper tropospheric jet in January 2016 is likely associated with the weakened 400 stratospheric polar vortex and the subsequent negative phase of the AO (Bell et al., 401 2009;Fletcher and Kushner, 2010), leading to more cold advection, enhanced EAWM, 402 and higher PBL height compared to the conditions during the peak of the El Niño with 403 lower PBL height (Fig. 8a). These changes in the meteorological conditions 404 associated with the changes in the El Niño development and AO phase shift and the 405 406 consequent changes in PM<sub>2.5</sub> 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 407 in the bottom of Fig. 9), the El Niño and positive AO (+AO) are in phase and weakens 408 the EAWM (with orange and blue arrows indicating effect from El Nino and AO, 409 respectively in Fig. 9a), leading to lower PBL and the subsequent severe haze 410 pollution. After the mature phase of El Niño (Fig. 9b), AO suddenly turns to the 411 negative phase (-AO), enhancing the EAWM (blue arrows in Fig. 9b) and subduing 412 the weakening effect from El Nino (the southerly orange arrow) on EAWM, resulting 413 414 in low PM<sub>2.5</sub> concentration.

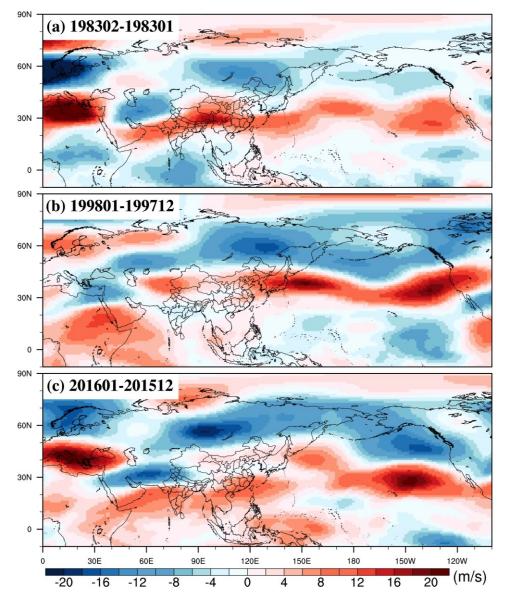


Fig. 7 Sub-seasonal changes of anomaly of 200 hPa zonal wind (relative to the
respective month in 1987-2016) during the start of the decay for three super El Niño
events, including the differences between anomaly of February 1983 and January
1983 (Fig. 7a), January 1998 and December 1997 (Fig. 7b), January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

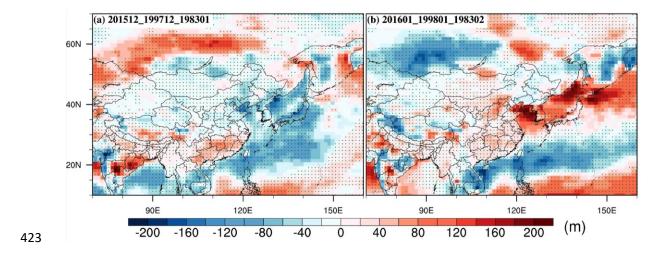


Fig. 8 Sub-seasonal changes of PBL height anomaly (relative to 1987-2016). The
anomaly was conducted over the respective month from 1987-2016, i.e., mean value
of anomaly over December 2015, December 1997 and January 1983 (Fig. 8a), and
January 2016, January 1998 and February 1983 (Fig. 8b). Stippled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

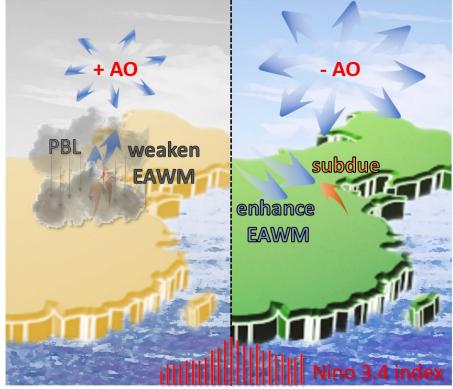


Fig. 9 Schematic of the modulation of El Niño and Arctic Oscillation on
atmospheric circulation and haze in northern China

#### 432 Summary and discussions

This study identifies a distinct seesaw feature of abnormally high and low monthly 433 434 mean PM<sub>2.5</sub> concentration in the two consecutive months of December 2015 and January 2016, respectively, in the North China Plain. Accompanying the seesaw 435 features of PM<sub>2.5</sub> are opposite large scale circulation patterns with positive (negative) 436 anomaly of 500 hPa geopotential height and southerly (northerly) wind anomalies at 437 the low level (850 hPa) over the North China Plain during December 2015 (January 438 2016). The contrast in PM2.5 between December 2015 and January 2016 is 439 significantly larger than the change from December to January between 2009 and 440 2017 recorded at US embassy Beijing. 441

442 As the modulation of meteorological conditions on PM<sub>2.5</sub> occurred during a strong El Niño period, we explored the relationship between strong El Niño and PM<sub>2.5</sub>. First, 443 using a regional climate/chemistry model WRF/CMAQ, we identified that all three 444 super El Niño events in the recent record (1982/83, 1997/98, and 2015/16) yield 445 similar seesaw modulation features of PM<sub>2.5</sub>. Further analysis showed that the seesaw 446 PM<sub>2.5</sub> variations are modulated by the combined effect of El Niño and Arctic 447 Oscillation (AO). In December 2015, the mature phase of an extreme El Niño, 448 accompanied by a positive AO, weakened the EAWM, as indicated by a positive 449 450 anomaly of geopotential height at mid-troposphere (i.e., 500 hPa) and southerly wind anomalies at the low level (i.e., 850 hPa) over the North China Plain (NCP), resulting 451 in reduced planetary boundary layer (PBL) height, abnormally warm temperature and 452 substantial haze accumulation during this period. In the following month (January 453 2016) when the El Niño began to decay, a sharp reversal of the AO from a positive 454 phase (in December 2015) to a negative phase triggered enhanced EAWM, inducing 455 cold advection and anomalous low-level northerly winds over the NCP that fostered 456 457 atmospheric dispersion and substantially reduced haze formation. This abrupt change of the AO from a positive to a negative phase was robustly found for the other two 458 extreme El Niño events during the corresponding decay period (February 1983 and 459

January 1998) after the mature phase (January 1983 and December 1997). These 460 changes in circulation are likely associated with a southward shift of the upper 461 troposphere jet at 200 hPa and weakened stratospheric vortex during the decay phase 462 compared to the mature phase of El Niño. As the frequency of super El Niño like the 463 2015/16 event is projected to increase in the future (Cai et al., 2014;Cai et al., 2015), 464 the seesaw modulation of super El Niño and AO may become more frequent, 465 revealing vital information useful for policy makers dealing with air quality issues in 466 467 China.

Although we performed WRF-CMAQ simulations to demonstrate the impacts of 468 atmospheric circulation during three super El Niño events on haze, this study did not 469 470 isolate the general effect of El Niño and AO on haze formation. To address this limitation, future studies will design Atmospheric Model Intercomparison Project 471 (AMIP) type scenarios, e.g., by running multi-ensemble members of scenarios with 472 global models such as Community Earth System Model (CESM) using different SSTs 473 474 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 475 and AO on haze formation. 476

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

Acknowledgement. This research was supported by grants from the National Natural Science
Foundation of China (91744208, 41705124), Shandong Provincial Natural Science Foundation,
China (ZR2017MD026) and the Fundamental Research Funds for the Central Universities
(201712006;201762010). PNNL is operated for DOE by Battelle Memorial Institute under
contract DE-AC05-76RL01830.

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## 484 **References**:

485 Baldwin, M. P., and Dunkerton, T. J.: Propagation of the Arctic Oscillation from the stratosphere to the

- 486 troposphere, J. Geophys. Res., 104, 30937-30946, 10.1029/1999JD900445, 1999.
- Bell, C. J., Gray, L. J., Charlton-Perez, A. J., Joshi, M. M., and Scaife, A. A.: Stratospheric Communication
  of El Niño Teleconnections to European Winter, J. Clim., 22, 4083-4096, 10.1175/2009JCLI2717.1,
  2009.
- Byun, D., and Ching, J. K. S.: Science Algorithms of the EPA Models-3 CommunityMultiscale Air Quality
  (CMAQ) Modeling System., U. S. Environmental Protection Agency, Office of Research and
  Development, EPA, Washington, DC, 727, 1999.
- Byun, D., and Schere, K. L.: Review of the governing equations, computational algorithms, and other
  components of the models-3 Community Multiscale Air Quality (CMAQ) modeling system,
  Applied Mechanics Reviews, 59, 51-77, 10.1115/1.2128636, 2006.
- Cai, W., Li, K., Liao, H., Wang, H., and Wu, L.: Weather conditions conducive to Beijing severe haze
  more frequent under climate change, Nature Clim. Change, 7, 257-262, 10.1038/nclimate3249,
  2017.
- Cai, W. J., Borlace, S., Lengaigne, M., van Rensch, P., Collins, M., Vecchi, G., Timmermann, A., Santoso,
  A., McPhaden, M. J., Wu, L. X., England, M. H., Wang, G. J., Guilyardi, E., and Jin, F. F.: Increasing
  frequency of extreme El Nino events due to greenhouse warming, Nature Climate Change, 4,
  111-116, 10.1038/nclimate2100, 2014.
- 503 Cai, W. J., Santoso, A., Wang, G. J., Yeh, S. W., An, S. I., Cobb, K. M., Collins, M., Guilyardi, E., Jin, F. F.,
- 504 Kug, J. S., Lengaigne, M., McPhaden, M. J., Takahashi, K., Timmermann, A., Vecchi, G., Watanabe,
- 505 M., and Wu, L. X.: ENSO and greenhouse warming, Nature Climate Change, 5, 849-859, 506 10.1038/nclimate2743, 2015.
- 507 Chang, J. S., Brost, R. A., Isaksen, I. S. A., Madronich, S., Middleton, P., Stockwell, W. R., and Walcek, C.
- J.: A three-dimensional Eulerian acid deposition model: Physical concepts and formulation, J.
  Geophys. Res., 92, 14681-14700, 10.1029/JD092iD12p14681, 1987.
- 510 Chang, L., Xu, J., Tie, X., and Wu, J.: Impact of the 2015 El Nino event on winter air quality in China, Sci.
- 511 Rep., 6, 34275, 10.1038/srep34275
- 512 https://www.nature.com/articles/srep34275#supplementary-information, 2016.
- 513 Chen, H., Lin, Y., Su, Q., and Cheng, L.: Spatial variation of multiple air pollutants and their potential

514 contributions to all-cause, respiratory, and cardiovascular mortality across China in 2015–2016,

515 Atmos. Environ., 168, 23-35, https://doi.org/10.1016/j.atmosenv.2017.09.006, 2017.

Cheng, X. H., Sun, Z. A., Li, D. P., Xu, X. D., Jia, M. W., and Cheng, S. Y.: Short-term aerosol radiative
effects and their regional difference during heavy haze episodes in January 2013 in China, Atmos.

518 Environ., 165, 248-263, 10.1016/j.atmosenv.2017.06.040, 2017.

Cheung, H. N., Zhou, W., Mok, H. Y., and Wu, M. C.: Relationship between Ural–Siberian Blocking and
the East Asian Winter Monsoon in Relation to the Arctic Oscillation and the El Niño–Southern

521 Oscillation, J. Clim., 25, 4242-4257, 10.1175/JCLI-D-11-00225.1, 2012.

- 522 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M.
- 523 A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N.,
- 524 Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E.
- 525 V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette,
- 526 J. J., Park, B. K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J. N., and Vitart, F.: The
- 527 ERA-Interim reanalysis: configuration and performance of the data assimilation system, Q. J. Roy.
  528 Meteor. Soc., 137, 553-597, 10.1002/qj.828, 2011.
- 529 Ding, A. J., Huang, X., Nie, W., Sun, J. N., Kerminen, V. M., Petäjä, T., Su, H., Cheng, Y. F., Yang, X. Q.,
- 530 Wang, M. H., Chi, X. G., Wang, J. P., Virkkula, A., Guo, W. D., Yuan, J., Wang, S. Y., Zhang, R. J., Wu, Y.
- 531 F., Song, Y., Zhu, T., Zilitinkevich, S., Kulmala, M., and Fu, C. B.: Enhanced haze pollution by black
- carbon in megacities in China, Geophys. Res. Lett., 43, 2873-2879, 10.1002/2016GL067745, 2016.
- Emery, C., Tai, E., and Yarwood, G.: Enhanced meteorological modeling and performance evaluation
  for two Texas episodes, Prepared for the Texas Natural Resource Conservation Commission, by
  ENVIRON International Corp, Novato, CA, 2001.
- Fletcher, C. G., and Kushner, P. J.: The Role of Linear Interference in the Annular Mode Response to
   Tropical SST Forcing, J. Clim., 24, 778-794, 10.1175/2010JCLI3735.1, 2010.
- 538 Gao, Y., Leung, L. R., Zhao, C., and Hagos, S.: Sensitivity of U.S. Summer Precipitation to Model
- 539 Resolution and Convective Parameterizations Across Gray Zone Resolutions, J. Geophys. Res., 122,
- 540 2714-2733, 10.1002/2016jd025896, 2017.
- 541 Geng, X., Zhang, W., Stuecker, M. F., and Jin, F.-F.: Strong sub-seasonal wintertime cooling over East
- 542 Asia and Northern Europe associated with super El Niño events, Sci. Rep., 7, 3770,

- 543 10.1038/s41598-017-03977-2, 2017.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global
  terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from
  Nature), Atmos Chem Phys, 6, 3181-3210, 2006.
- Han, F., Xu, J., He, Y., Dang, H., Yang, X., and Meng, F.: Vertical structure of foggy haze over the
  Beijing-Tianjin-Hebei area in January 2013, Atmos. Environ., 139, 192-204,
  10.1016/j.atmosenv.2016.05.030, 2016.
- He, J., Gong, S., Yu, Y., Yu, L., Wu, L., Mao, H., Song, C., Zhao, S., Liu, H., Li, X., and Li, R.: Air pollution
  characteristics and their relation to meteorological conditions during 2014–2015 in major Chinese
  cities, Environ. Pollut., 223, 484-496, <u>https://doi.org/10.1016/j.envpol.2017.01.050</u>, 2017.
- Hui, G., and Xiang, L.: Influences of El Nino Southern Oscillation events on haze frequency in eastern
  China during boreal winters, Int. J. Climatol., 35, 2682-2688, 10.1002/joc.4133, 2015.
- Janjić, Z. I.: The Step-Mountain Coordinate: Physical Package, Mon. Weather. Rev., 118, 1429-1443,

556 10.1175/1520-0493(1990)118<1429:tsmcpp>2.0.co;2, 1990.

- Jeong, J. I., and Park, R. J.: Winter monsoon variability and its impact on aerosol concentrations in East
   Asia, Environ. Pollut., 221, 285-292, https://doi.org/10.1016/j.envpol.2016.11.075, 2017.
- 559 Kajino, M., Ueda, H., Han, Z., Rei, K., Inomata, Y., and Kaku, H.: Synergy between air pollution and 560 urban meteorological changes through aerosol-radiation-diffusion feedback A case study of
- 561 Beijing in January 2013, Atmos. Environ., 171, 98-110, 10.10164/j.atmosenv.2017.10.018, 2017.
- 562 Kwok, R. H. F., Napelenok, S. L., and Baker, K. R.: Implementation and evaluation of PM2.5 source
  563 contribution analysis in a photochemical model, Atmos. Environ., 80, 398-407,
  564 https://doi.org/10.1016/j.atmosenv.2013.08.017, 2013.
- Li, K., Liao, H., Cai, W., and Yang, Y.: Attribution of Anthropogenic Influence on Atmospheric Patterns
  Conducive to Recent Most Severe Haze Over Eastern China, Geophys. Res. Lett., in press.,
  10.1002/2017GL076570, 2018.
- Li, M., Liu, H., Geng, G., Hong, C., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H., Man, H., Zhang, Q., and
  He, K.: Anthropogenic emission inventories in China: a review, National Science Review, 4,
  834-866, 10.1093/nsr/nwx150, 2017.
- 571 Li, Q., Zhang, R., and Wang, Y.: Interannual variation of the wintertime fog-haze days across central

- and eastern China and its relation with East Asian winter monsoon, Int. J. Climatol., 36,
  346-354, 10.1002/joc.4350, 2016.
- Liang, F., Tian, L., Guo, Q., Westerdahl, D., Liu, Y., Jin, X., Li, G., and Pan, X.: Associations of PM2.5 and
  Black Carbon with Hospital Emergency Room Visits during Heavy Haze Events: A Case Study in
  Beijing, China, Int. J. Environ. Res. Public Health, 14, 725, 2017.
- Liu, F., Zhang, Q., A., R. J. v. d., Zheng, B., Tong, D., Yan, L., Zheng, Y., and He, K.: Recent reduction in NO
   x emissions over China: synthesis of satellite observations and emission inventories, Environ. Res.
- 579 Lett., 11, 114002, 2016.
- Liu, Q., Sheng, L., Cao, Z., Diao, Y., Wang, W., and Zhou, Y.: Dual effects of the winter monsoon on
  haze-fog variations in eastern China, J. Geophys. Res., 122, 5857-5869, 10.1002/2016JD026296,
  2017.
- Ma, M., Gao, Y., Zhang, T., Wang, Y., Zhang, S., Leung, L. R., Liu, C., Wang, S., Zhao, B., Chang, X., Su, H.,
  Yao, X., and Gao, H.: The substantial contribution of land use change and heat waves on the
  recent high ozone events in North China, in preparation.
- 586 Mellor, G. L., and Yamada, T.: DEVELOPMENT OF A TURBULENCE CLOSURE-MODEL FOR GEOPHYSICAL
   587 FLUID PROBLEMS, Reviews of Geophysics, 20, 851-875, 1982.
- Nie, Y., Sun, L., Wang, D., and Li, D.: Possible Causes for the Sudden Drop of Air Temperature in the
   Northern Hemisphere from Early- to Mid-Winter, Meteorological Monthly, 42, 1223-1229, 2016.
- 590 Otte, T. L., and Pleim, J. E.: The Meteorology-Chemistry Interface Processor (MCIP) for the CMAQ
- 591 modeling system: updates through MCIPv3.4.1, Geosci. Model Dev., 3, 243-256,
  592 10.5194/gmd-3-243-2010, 2010.
- 593 Pleim, J., and Ran, L.: Surface Flux Modeling for Air Quality Applications, Atmosphere, 2, 271-302,
  594 doi:10.3390/atmos2030271, 2011.
- Qin, Y., and Xie, S. D.: Historical estimation of carbonaceous aerosol emissions from biomass open
   burning in China for the period 1990–2005, Environ. Pollut., 159, 3316-3323,
   <u>https://doi.org/10.1016/j.envpol.2011.08.042</u>, 2011.
- Rayner, N. A., Parker, D. E., Horton, E. B., Folland, C. K., Alexander, L. V., Rowell, D. P., Kent, E. C., and
  Kaplan, A.: Global analyses of sea surface temperature, sea ice, and night marine air temperature
- since the late nineteenth century, J. Geophys. Res., 108, n/a-n/a, 10.1029/2002JD002670, 2003.

- 601 Ren, H. L., Wang, R., Zhai, P., Ding, Y., and Bo, L. U.: Upper-Ocean Dynamical Features and Prediction of
- the Super El Nino in 2015/16: A Comparison with the Cases in 1982/83 and 1997/98, Journal of
  Meteorological Research, 31, 278-294, 2017.
- 504 Saha, S., Moorthi, S., Wu, X., Wang, J., Nadiga, S., Tripp, P., Behringer, D., Hou, Y.-T., Chuang, H.-y.,

605 Iredell, M., Ek, M., Meng, J., Yang, R., Mendez, M. P., van den Dool, H., Zhang, Q., Wang, W., Chen,

606 M., and Becker, E.: The NCEP Climate Forecast System Version 2, J. Clim., 27, 2185-2208,

607 10.1175/JCLI-D-12-00823.1, 2013.

- Shi, Z., Li, J., Huang, L., Wang, P., Wu, L., Ying, Q., Zhang, H., Lu, L., Liu, X., Liao, H., and Hu, J.: Source
  apportionment of fine particulate matter in China in 2013 using a source-oriented chemical
  transport model, Sci. Total Environ., 601, 1476-1487, 10.1016/j.scitotenv.2017.06.019, 2017.
- 611 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X., Wang, W.,
- and Powers, J. G.: A description of the advanced research WRF version 3, NCAR Tech. Note,
  NCAR/TN-475+STR, 8 pp., Natl. Cent. for Atmos. Res., Boulder, CO, USA, available at:
  http://www.mmm.ucar. edu/wrf/users/docs/arw v3.pdf, 2008, 2008.
- 615 Song, C., Wu, L., Xie, Y., He, J., Chen, X., Wang, T., Lin, Y., Jin, T., Wang, A., Liu, Y., Dai, Q., Liu, B., Wang,
- 616 Y.-n., and Mao, H.: Air pollution in China: Status and spatiotemporal variations, Environ. Pollut.,

617 227, 334-347, <u>https://doi.org/10.1016/j.envpol.2017.04.075</u>, 2017.

- 518 Tie, X., Huang, R.-J., Cao, J., Zhang, Q., Cheng, Y., Su, H., Chang, D., Pöschl, U., Hoffmann, T., Dusek, U.,
- 619 Li, G., Worsnop, D. R., and O'Dowd, C. D.: Severe Pollution in China Amplified by Atmospheric
- 620 Moisture, Sci. Rep., 7, 15760, 10.1038/s41598-017-15909-1, 2017.
- USEPA: Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air
   Quality Goals for Ozone, PM2.5. and Regional Haze, EPA-454/B-07e002, 2007.
- 623 Wang, J., Zhao, B., Wang, S., Yang, F., Xing, J., Morawska, L., Ding, A., Kulmala, M., Kerminen, V.-M.,
- 624 Kujansuu, J., Wang, Z., Ding, D., Zhang, X., Wang, H., Tian, M., Petäjä, T., Jiang, J., and Hao, J.:
- 625 Particulate matter pollution over China and the effects of control policies, Sci. Total Environ.,
- 626 584-585, 426-447, <u>https://doi.org/10.1016/j.scitotenv.2017.01.027</u>, 2017.
- Wang, S. X., and Zhang, C. Y.: Spatial and temporal distribution of air pollutant emissions from open
  burning of crop residues in China, Sciencepaper Onlin, 3, 1-6, 2008.
- 629 Wang, S. X., Zhao, B., Cai, S. Y., Klimont, Z., Nielsen, C. P., Morikawa, T., Woo, J. H., Kim, Y., Fu, X., Xu, J.

- Y., Hao, J. M., and He, K. B.: Emission trends and mitigation options for air pollutants in East Asia,
  Atmos Chem Phys, 14, 6571-6603, DOI 10.5194/acp-14-6571-2014, 2014.
- Wu, G. X., Li, Z. Q., Fu, C. B., Zhang, X. Y., Zhang, R. Y., Zhang, R. H., Zhou, T. J., Li, J. P., Li, J. D., Zhou, D.
  G., Wu, L., Zhou, L. T., He, B., and Huang, R. H.: Advances in studying interactions between
  aerosols and monsoon in China, Science China: Earth Sciences, 45, 1609-1627,
  10.1007/s11430-015-5198-z, 2015.
- Ku, J., Chang, L., Yan, F., and He, J.: Role of climate anomalies on decadal variation in the occurrence of
   wintertime haze in the Yangtze River Delta, China, Sci. Total Environ., 599-600, 918-925,
   <a href="https://doi.org/10.1016/j.scitotenv.2017.05.015">https://doi.org/10.1016/j.scitotenv.2017.05.015</a>, 2017a.
- Ku, L., Yu, J.-Y., Schnell, J. L., and Prather, M. J.: The seasonality and geographic dependence of ENSO
  impacts on U.S. surface ozone variability, Geophys. Res. Lett., 44, 3420-3428,
  10.1002/2017GL073044, 2017b.
- Xu, Q., Wang, S., Guo, Y., Wang, C., Huang, F., Li, X., Gao, Q., Wu, L., Tao, L., Guo, J., Wang, W., and Guo,
  X.: Acute exposure to fine particulate matter and cardiovascular hospital emergency room visits in
  Beijing, China, Environ. Pollut., 220, 317-327, <u>https://doi.org/10.1016/j.envpol.2016.09.065</u>,
  2017c.
- Xue, F., Dong, X., and Fan, F. X.: Anomalous Western Pacific Subtropical High during El Nio Developing
  Summer in Comparison with Decaying Summer, Adv. Atmos. Sci., 35, 360-367,
  10.1007/s00376-017-7046-x, 2018.
- Kue, Y., and Kumar, A.: Evolution of the 2015/16 El Niño and historical perspective since 1979, SCIENCE
  CHINA Earth Sciences, 60, 1572, doi:<u>https://doi.org/10.1007/s11430-016-0106-9</u>, 2017.
- Yang, Y., Liao, H., and Lou, S.: Increase in winter haze over eastern China in recent decades: Roles of
  variations in meteorological parameters and anthropogenic emissions, J. Geophys. Res., 121,
  13,050-013,065, 10.1002/2016JD025136, 2016.
- 654 Ye, X., Song, Y., Cai, X., and Zhang, H.: Study on the synoptic flow patterns and boundary layer process
- of the severe haze events over the North China Plain in January 2013, Atmos. Environ., 124, 129-145, 10.1016/j.atmosenv.2015.06.011, 2016.
- 657 Yin, Z., Wang, H., and Chen, H.: Understanding severe winter haze events in the North China Plain in
- 658 2014: roles of climate anomalies, Atmos. Chem. Phys., 17, 1641-1651, 10.5194/acp-17-1641-2017,

659 2017.

- Yuan, Y., and Yang, S.: Impacts of Different Types of El Nino on the East Asian Climate: Focus on ENSO
  Cycles, J. Clim., 25, 7702-7722, 10.1175/jcli-d-11-00576.1, 2012.
- 662 Yuan, Y., Zhou, N., and Li, C.: Correlation between haze in North China and super El Niño events.,

663 Chinese J. Geophys, 60, 11-20, 10.6038/cjg20170102, 2017.

- 264 Zhang, Q., Quan, J., Tie, X., Li, X., Liu, Q., Gao, Y., and Zhao, D.: Effects of meteorology and secondary
- particle formation on visibility during heavy haze events in Beijing, China, Sci. Total Environ., 502,
  578-584, <u>http://dx.doi.org/10.1016/j.scitotenv.2014.09.079</u>, 2015.
- 267 Zhang, Y., Ding, A., Mao, H., Nie, W., Zhou, D., Liu, L., Huang, X., and Fu, C.: Impact of synoptic weather

668 patterns and inter-decadal climate variability on air quality in the North China Plain during 1980–

- 669 2013, Atmos. Environ., 124, 119-128, <u>https://doi.org/10.1016/j.atmosenv.2015.05.063</u>, 2016.
- Zhang, Z., Gong, D., Mao, R., Kim, S.-J., Xu, J., Zhao, X., and Ma, Z.: Cause and predictability for the
  severe haze pollution in downtown Beijing in November–December 2015, Sci. Total Environ., 592,

672 627-638, <u>https://doi.org/10.1016/j.scitotenv.2017.03.009</u>, 2017.

Thao, B., Wang, S. X., Liu, H., Xu, J. Y., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.:

NOx emissions in China: historical trends and future perspectives, Atmos. Chem. Phys., 13,
9869-9897, 10.5194/acp-13-9869-2013, 2013.

- 676 Zhao, B., Wu, W. J., Wang, S. X., Xing, J., Chang, X., Liou, K. N., Jiang, J. H., Gu, Y., Jang, C., Fu, J. S., Zhu,
- 677 Y., Wang, J. D., Lin, Y., and Hao, J. M.: A modeling study of the nonlinear response of fine particles
- to air pollutant emissions in the Beijing-Tianjin-Hebei region, Atmos Chem Phys, 17, 12031-12050,
- 679 DOI 10.5194/acp-17-12031-2017, 2017.
- 680 Zhao, B., Zheng, H., Wang, S., Smith, K. R., Lu, X., Aunan, K., Gu, Y., Wang, Y., Ding, D., Xing, J., Fu, X.,
- 681 Yang, X., Liou, K.-N., and Hao, J.: Change in household fuels dominates the decrease in PM2.5
- exposure and premature mortality in China in 2005-2015, Proceedings of the National Academy
- of Sciences of the United States of America, 10.1073/pnas.1812955115, 2018.
- Zhao, S., Li, J., and Sun, C.: Decadal variability in the occurrence of wintertime haze in central eastern
  China tied to the Pacific Decadal Oscillation, Sci. Rep., 6, 27424, 10.1038/srep27424, 2016.