Comments from Review 1:

This paper analyzes a specific haze event in North China, which features high and low PM2.5 concentrations in adjacent two months (December 2015 and January 2016). The seesaw pattern is modulated by circulation patterns related to a super El Nino event and a phase change of the Arctic Oscillation from positive to negative. This is supported by both observations and model results. The authors also conduct some additional model simulations and show similar seesaw features of PM2.5 concentrations during other super El Nino periods (1997-1998 and 1982-1983). The manuscript is scientifically correct and the results are well laid out. My major concern is that the role of removal in affecting PM2.5 concentrations is neglected in the manuscript. There are some points that I hope the authors could discuss or clarify in the revision.

Response: We thank the reviewer for the constructive comments to help us further improve the manuscript. Please see the detailed responses to your comments below.

1、The removal of $PM_{2.5}$, such as wet and dry deposition, is not discussed in the paper. How are these processes represented in the model? Could they also play a role in the seesaw pattern of $PM_{2.5}$ concentrations?

Response: We have added the descriptions about dry and wet deposition in the third paragraph of section 2 in the revised manuscript. The descriptions were also shown below.

"The removal of particulate matter includes dry and wet deposition. For dry deposition, it is in general expressed by the product of dry deposition velocity and concentration of pollutants. The dry deposition velocity is the inverse of resistance including aerodynamic resistance, molecular motion and surface resistance, with more details in Pleim and Ran (2011). The wet deposition of pollutants depends on their concentrations in cloud water and the precipitation rate, and the algorithms were based on regional acid deposition model (RADM; (Chang et al., 1987))."

In order to elucidate the role of depositions in the removal of $PM_{2.5}$, we have re-run the simulations for December 2015 and January 2016 by adding the process analysis. A total of nine process are included, including horizontal and vertical advection/diffusion, dry and wet deposition, aerosol chemistry, gas phase chemistry and emissions. We found that during these two months, the wet deposition plays little role due to small amount of precipitation, and the dry deposition accounts for about 10% (12% December 2015 and 13% January 2016). The comparable contributions from the deposition in December 2015 and January 2016 indicates that the seesaw patterns were not related too much with the deposition. These points have been reflected in the last paragraph of section 4.3 in the revised manuscript.

Pleim, J., and Ran, L.: Surface Flux Modeling for Air Quality Applications, Atmosphere, 2, 271-302, doi:10.3390/atmos2030271, 2011.
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.

2、I wonder why the authors compare the difference between anomalies of PM2.5 concentrations rather than the difference between absolute values throughout the paper(except for Fig.7 if I understand it correctly)? I don't think the conclusion would change much if absolute values were used. If the authors decide to use anomalies, some description or figures of climatological values might be helpful.
Response: The anomalies of difference were used to eliminate the effect of climatological mean. As the reviewer suggested, we have added the climatological values of $PM_{2.5}$ in Figures S1 in the supporting information.

3、A more detailed discussion on Fig. 10, in the main text or in the caption, would be helpful. Currently it is not very clear what those arrows in Fig. 10 indicate.

Response: We have added more detailed discussion of this figure (Fig. 9 in the revised manuscript) in the last paragraph of section 4.4.

Technical corrections:
1. The unit for wind vector in Fig.3 and Fig. 4 is missing.

Response: The unit has been added in the revised caption.

2. It might be helpful to show the NCP box in the figures of meteorological conditions (e.g., Figs. 3,4,5) as well.

Response: We tried to add the NCP box in Figures 3,4,5, but found that the box was redundant since Figure 1 has already showed it. Thus, we removed the box.

3. Lines 254-271：In this paragraph, (Fig. 5, Fig. 6) should be (Fig. 4, Fig. 5). Fig. 5 may seem somewhat redundant, as near surface wind anomaly has similar pattern as 850 hPa wind anomaly and does not seem to provide additional information.

Response: The typo was corrected. Figure 5 has been moved to the supporting information based on the reviewer's suggestion.

Comments from Review 2:

This paper described distinct pollution levels in two consecutive months of winter 2015 in North China. and argued that such a feature is regulated by meteorological patterns connected to the El Nino and Arctic Oscillations (AO). The study uses observations of one single super El Nino year to raise the hypothesis on ENSO/AO influence on haze distribution. The study then used WRF model simulations of three other super El Nino years to test those hypotheses. In terms of mechanism, this paper claims that the combination effect of El Nino and AO can influence the intensity of EAWM and thus result in an anomalous PM2.5 levels. I found the overall presentation straightforward and the topic is within the scope of ACP. However, the following concerns shall be addressed before its publication.

Response: We thank the reviewer for the comprehensive evaluation and comments to help to improve the manuscript. Please see the detailed responses to your comments below.

1. This paper states the importance of boundary layer height several times without mentioning the schemes and calculation used in the model. Please clarify.

Response: The PBL scheme of Mellor-Yamada-Janjic was used and added in the revised manuscript.

2. The simulation domain was not specified at all. It says "...with physics options same as those discussed in Gao et al. (2017)...". However, in Gao et al. (2017), the simulation was conducted in U.S. but this study is done for East Asia. Please specify the basic domain information.

Response: The domain information was added in the revised manuscript (the third paragraph of section 2), which is also shown below.

"The domain covers majority of East Asia (shown latter; i.e., Fig. 5b), with spatial resolution of 36 km by 36 km. The pressure of the model top is 50 hPa, with lambert conformal conic projection centered at 34°N, 110°E. A total of 34 layers were used, with the top of the first layer at about 40 meters."

3. For biomass burning, the paper states "...biomass burning emissions include open burning of agricultural residue, calculated based on crop yields, fraction of biomass burned in the open field...". But the paper did not specify the inventory used in this case. So which inventory did you use? FINN, GFED or any other self-developed source specified for this region?

Response: In this study, we used self-developed the emission inventory, developed by Tsinghua University. The details have been added in the revised manuscript (last paragraph of section 2), which is also shown below:

“The emissions from open burning of agricultural residue have been included in the anthropogenic emission inventory developed by Tsinghua University. They were calculated based on crop yields, the 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).”

4.The length of spin-up time is questionable. In this paper, the spin-up time for each simulation is only one week. Kumar et al. (2013) shows that it takes WRF-Chem about 10 days to spin-up for free atmosphere and 20 days to spin-up for surface level.

Response: Regarding the spin-up period, we have done a few tests previously, and found no significant differences once the spinning-off period reaches a week or longer. In some other studies, even fewer spinning-off period was used, i.e., Im et al. (2010) used 3-day and Sartelet et al. (2007) used one-day as spin-up period. Therefore, the one-week spin-up time should be sufficient.

Im, U., Markakis, K., Unal, A., Kindap, T., Poupkou, A., Incecik, S., Yenigun, O., Melas, D., Theodosi, C., and Mihalopoulos, N.: Study of a winter PM episode in Istanbul using the high resolution WRF/CMAQ modeling system, Atmos. Environ., 44, 3085-3094, https://doi.org/10.1016/j.atmosenv.2010.05.036, 2010.

Sartelet, K. N, H. Hayami and B. Sportisse. Dominant aerosol processes during high-pollution episodes over Greater Tokyo. Journal of Geophysical Research: Atmospheres, 2007, 112(D14).

5.Fig. 3, 4 and 5 all stated that “Stippled areas indicating exceedance of 90th confidence interval.” But what kind of statistical test was applied here? Also what are the samples?

Response: The 90th confidence interval was calculated for each grid separately based on a two tailed t-distribution method using the thirty-year (1987-2016) data. The sample size is 30. For example, for December 2015 (Fig. 3) in terms of geopotential height (GHT) at 500 hPa, 90th confidence interval was calculated using GHT from December 1987-2016. Then, the value in December 2015 for each grid was compared to the 90th confidence interval. This has been reflected in the caption of Figure 3 in the revised manuscript.

6.The last paragraph on future research is also a bit puzzling, running SST forced experiment can be helpful for ENSO, but AO is difficult to be forced by SST.

Response:

The reviewer is correct that SST is directly related to ENSO. However, as was discussed in Geng et al. (2017) as well as our manuscript, we found that during the super El Niño events including 1982/83, 1997/98, and 2015/16, in particular during the peak of El Niño period, it was accompanied by a rapid sub-seasonal North Atlantic Oscillation (NAO)/Arctic Oscillation (AO) phase reversal from a positive to negative phase. The ENSO and AO is interconnected in this regard. Therefore, we propose to conduct a SST experiment to further elucidate the mechanism and the subsequent impact on haze formation.

Geng, X., Zhang, W., Stuecker, M. F., and Jin, F.-F.: Strong sub-seasonal wintertime cooling over East Asia and Northern Europe associated with super El Niño events, Sci. Rep., 7, 3770, 10.1038/s41598-017-03977-2, 2017.

1. Please consider to replace the sequential color schemes with divergent color schemes when showing the anomalies (e.g., Fig.1 and Fig.7). Fig 3 is a better example.

Response: The color schemes have been changed based on the reviewer's suggestions.

2. When drawing the boundaries of NCP in Fig.1, please be more rigorous. The box area is not entirely NCP. It also contains part of Bohai Sea and Inner Mongolia Plateau. Although this may not affect your final results but can cause misleading when saying this is NCP.

Response: The box does not include Inner Mongolia (Fig. 1 of the revised manuscript), and we added the relevant descriptions, i.e., the exclusion of Bohai area.

3. What is the unit of the wind vector in Fig. 3? m/s?

Response: The unit of m/s has been added in the caption of Fig. 3.

4. The captions of this paper need to be clearer. There are lots of figures with sub panels (e.g., a,b etc.) but they are not specifically mentioned in the caption. This way of description can be very confusing.

Response: All captions have been checked and made clearer by adding more specific descriptions.

5. Line 204-205: "the emissions of SO_2 in January is usually higher than December primarily due to a higher power demand" this statement needs a reference.

Response: We have rephrased the sentence and SO_2 emission was inferred by the concentration. The revised descriptions (last paragraph of section 3.2) are shown below:

Moreover, the anthropogenic emissions in January could be comparable to or higher than that in December, i.e., in January 2016, higher SO_2 concentration, implicative of SO_2 emissions, was found than December 2015 based on observed data (http://www.pm25.in; not shown).

6. Fig. 6b shows very low PM2.5 concentrations for major cities other than China and India. Why is this the case? How about cities like Tokyo, Osaka, Seoul and Bangkok etc.?

Response: Based on the data available to us, we added the evaluation of 13 stations in Japan, South Korea and Thailand. The observational data is from EANET (Acid Deposition Monitoring Network in East Asia, http://www.eanet.asia/product/index.html). The concentrations in these cities are lower than that from the major cities in China. Please note over Cheju and Kanghwa in South Korea, only the data in January 2016 is available. From the scatter plots shown below, we can tell that the model performs well (with low mean bias and error) among these stations. The descriptions as well as the figure and table have been added to the part 2 of the supporting information.

Table S1. Station information of EANET sites

	Japan										South Korea		Thailand
Stations	Rishiri	Ochiishi	Tappi	Sado-seki	Happo	Ijira	Oki	Banryu	Yusuhara	Hedo	Cheju	Kanghwa	Bankok
Latitude	45.12	43.20	41.25	38.25	36.68	35.57	36.28	34.67	32.73	26.78	33.52	37.74	13.75
Longitude	141.23	145.52	141.35	138.40	137.80	136.70	133.18	131.70	132.98	128.23	126.52	126.49	100.50

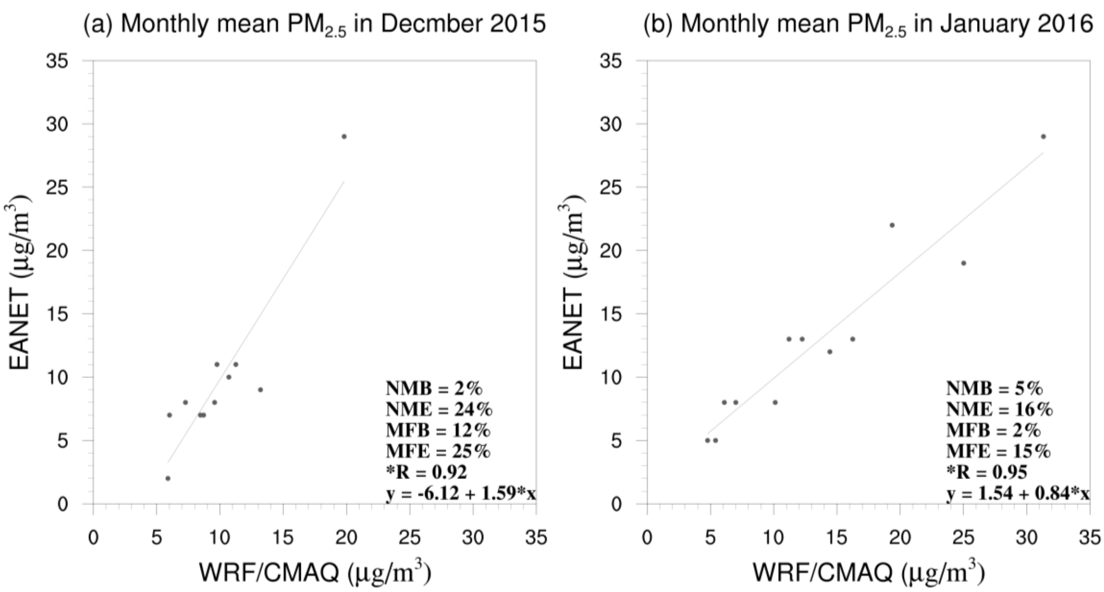

Figure S9 Evaluation of monthly $PM_{2.5}$ in CMAQ based on selected EANET observational data: (NMB: Normalized Mean Bias; NME: Normalized Mean Error;

MFB: Mean Fractional Bias; MFE: Mean Fractional Error; R: correlation coefficient). The statistical significance of the linear correlation coefficient was performed and *R implies statistical significance at the 95% confidence level.

7. In Table 1, can you explain the large bias of 201512 WD10? It is almost 20 degrees and should not be treat as negligible.

Response: slightly larger bias (19.25°) of wind direction at 10-m (WD10) in December 2015 is partly attributable to the model comparison with observed values close to 0° or 360°, which may yield large bias albeit the small differences in reality (i.e., 10° in model vs. 350° in observation). This has been added in section 4.3 in the revised manuscript.

8. Line 62. “Formation” should be distribution. A relevant reference is Chen et al., (2018).

Response: “Formation” has been changed to “distribution”.

Comments from Review 3:

Response: We thank the reviewer for the thorough comments to improve our manuscript. Regarding the reviewer’s concern, we have addressed all the comments shown below.

The authors identified an interesting sub-seasonal seesaw pattern of winter haze pollution in northern China, featuring high and low PM2.5 concentrations in two adjacent months. They also found that this phenomenon is related to the circulation patterns modulated by El Nino and Arctic Oscillations. In general, I think this manuscript is well structured and the topic is suitable for ACP. But I still have some concerns about the robustness of the proposed mechanism before ACP accepts this paper.

1、The analysis is based on only three super El Nino events after the 1980s. The number of cases is too few here. Does the proposed mechanism also apply to the El Nino events with smaller magnitudes and these before the 1980s? I think the readers also would like to see a figure displaying the circulation anomalies from an ensemble of El Nino events.

Response: Based on reviewer’s suggestion, we now have checked the circulation patterns (i.e., 500 hPa GHT) during all the El Nino events since 1948, and found that there were no obvious seesaw patterns in the other El Nino events. The reason we selected 1948 is that the reanalysis data was only available from 1948. Only the three El Nino events (1982/1983, /1997/1998 and 2015/2016) were super strong El Nino events (peak of 3-month running SST greater than 2.0 °C), and all the other events belong to moderate or weak events. As the reviewer suggested, we added the circulation anomalies from an ensemble of El Nino events, including the super El Nino events, the moderate/small events after 1980 and the events from 1948-1980. The peak and decay of the ensemble of El Nino events were shown in Fig. S5a-f in the supporting information, which were also shown below. The results indicate that only the super El Nino events shows obvious seesaw patterns, whereas the ensembles of the others events do not show the seesaw patterns, which further implies the robustness of the seesaw patterns we discussed. This has been added in the revised manuscript (first paragraph of section 4.2).

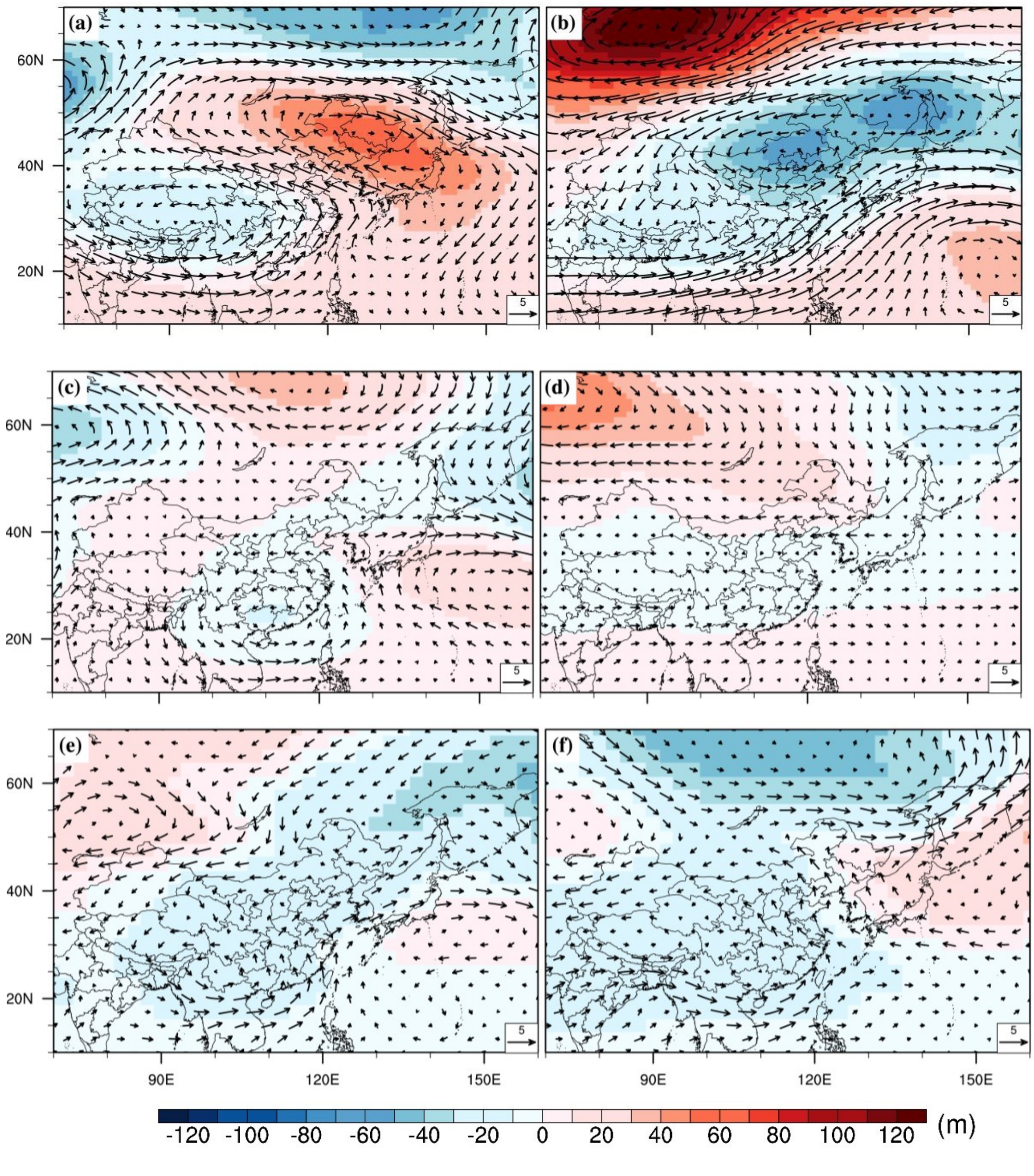

Fig. S5. Anomaly (relative to 1987-2016) of geopotential height and wind vector at 500 hPa for the ensemble mean of El Nino events, i.e., Fig. S5a (peak of super El Nino: 198301/199712/201512), Fig. S5b (the decay of super El Nino: 198302/199801/201601), Fig. S5c (peak of moderate El Nino events since 1980:

199201/199412/200211/200412/200612/200912), Fig. S5d (the decay of moderate El Nino events since 1980: 199202/199501/200212/200501/200701/201001), Fig. S5e (peak of El Nino events before 1980: 195801/196311/196511/197211/197711), Fig. S5f (decay of El Nino events before 1980: 195802/196312/196512/197212/197712)

2、If the El Nino peaks in December 2015, its effects on northern China winds may appear one or two months later due to the time spent on the wave propagation. So I am wondering whether the El Nino really causes the high $PM_{2.5}$ concentrations in December 2015. I wish the authors can have some comments here.

Response: We thank the reviewer for raising the concern regarding the impact from El Nino. The effect of El Nino on haze formation was mainly through the modulation on EAWM. Since the three month running mean SST was used, the peak of El Nino in a certain degree has reflected a lag of one month or so. In addition, as was discussed in the conclusions, the mature phase of super El Nino was accompanied by a positive AO, and these two factors together modulate the changes of EAWM and the subsequent haze formation. Therefore, we are confident of the effect of El Nino on haze formation.

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

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Abstract

Utilizing a recent observational particulate matter with diameters less than 2.5 µm ($PM_{2.5}$) dataset in North China, this study reveals a distinct seesaw feature of abnormally high and low $PM_{2.5}$ concentrations in the adjacent two months of December 2015 and January 2016, accompanied by distinct meteorological modulations. The seesaw pattern is postulated to be linked to super El Niño and the Arctic Oscillation (AO). During the mature phase of El Niño in December 2015, the weakened East Asian Winter Monsoon (EAWM) and the associated low-level southerly wind anomaly reduced planetary boundary layer height, favoring strong haze formation. This circulation pattern was completely reversed in the following 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 upper-tropospheric jet from December to January relative to climatology, leading to an enhanced EAWM and substantially lower haze formation. This sub-seasonal 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. Numerical experiments with the Weather Research and Forecasting (WRF)-Community Multi-scale Air Quality (CMAQ) model were used to test the modulation of the meteorological conditions on haze formation. With the same emission, simulations for three super El Niño periods (1983, 1997 and 2015) robustly show higher $PM_{2.5}$ concentrations under the mature phase of the super El Niño, but 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 circulation anomaly on $PM_{2.5}$ concentrations in North China.

Key words: Haze, El Niño, Arctic Oscillation, East Asian winter monsoon, WRF/CMAQ

1. Introduction

China has experienced severe haze pollution in recent years (Ding et al., 2016;Song et al., 2017;Tie et al., 2017;Zhang et al., 2015), with potentially detrimental effect on human health (Chen et al., 2017) as suggested by a substantial increase in hospital visits during the haze season (Liang et al., 2017;Xu et al., 2017c). Understanding the mechanism of haze formation is vital for developing effective measures to relieve the haze pollution. Despite the continued reduction in anthropogenic emission such as NO_x, SO_2, and CO in China in the past few years (Li et al., 2017;Liu et al., 2016;Wang et al., 2017), the severe haze pollution motivates a need to understand the mechanism of haze ~~formation~~ distribution from a meteorological perspective (Ding et al., 2016;He et al., 2017). A few studies investigated the relationship between haze and climate variability at decadal or longer time scales (Jeong and Park, 2017;Liu et al., 2017;Wu et al., 2015;Xu et al., 2017a;Yin et al., 2017;Zhang et al., 2016), while others examined the meteorological factors associated 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) found that the number of fog–haze days in winter across central and eastern China has a strong relationship with the East Asian winter monsoon (EAWM), with weak 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 simulation from 1985 to 2005 with the Goddard Earth-Observing System chemical transport model (GEOS-Chem). Zooming in on the severe haze event in 2013, Cai et al. (2017) identified the conducive weather conditions for severe haze including weakened surface northerlies and northward shift of mid-troposphere northwesterlies extending to the north of Beijing, reducing the cold and dry flow to Beijing.

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

While it is highly possible that the severe haze pollution in November to December of 2015 is a result of the strong El Niño, the sudden drop in $PM_{2.5}$ concentrations in January 2016 compared to December 2015 has heretofore largely been ignored. Puzzled by the seesaw pattern of severe haze pollution in December 2015 and the low $PM_{2.5}$ concentrations in January 2016, this study aims to explore the possible mechanism behind this temporal seesaw phenomenon. The paper is organized as follows. In Section 2, we introduce the data and methodology used in the study, followed by the monthly variations of $PM_{2.5}$ concentration. In Section 4, mechanisms regarding the haze variations are investigated.

2. Data and methodology

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 l.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 $PM_{2.5}$ 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 l.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) (Skamarock et al., 2008) model, coupled to a chemistry model, Community Multi-scale Air Quality (CMAQ), was used to investigate possible factors modulating haze formation. WRF version 3.8.1 was used in this study, with physics options same as those discussed in Gao et al. (2017), i.e., for planetary boundary layer (PBL), the Mellor-Yamada-Janjic scheme was used (Janjić, 1990;Mellor and Yamada, 1982). The domain covers majority of East Asia (shown lat~~t~~er; i.e., Fig. 5b), with spatial resolution of 36 km by 36 km. The pressure of the model top is 50 hPa, with lambert conformal conic projection centered at 34°N, 110°E. A total of 34 layers were used, with the top of the first layer at about 40 meters. The initial and boundary conditions for WRF were provided by the NCEP Climate Forecast System Reanalysis (CFSR) version 2 (Saha et al., 2013), with a spatial resolution of 0.5 deg×0.5 deg. For regional chemistry, the widely used CMAQ model (Byun and Ching, 1999;Byun and Schere, 2006), with the latest version 5.2, was used in this study, with the carbon-bond version

6 (CB06) for the major gas phase chemistry and AERO6 for aerosol module. The removal of particulate matter includes dry and wet deposition. For dry deposition, it is in general expressed by the product of dry deposition velocity and concentration of pollutants. The dry deposition velocity is the inverse of resistance including aerodynamic resistance, molecular motion and surface resistance, and more details were discussed in Pleim and Ran (2011). The wet deposition of pollutants depends on their concentrations in cloud water and the precipitation rate, and the algorithms were based on regional acid deposition model (RADM; (Chang et al., 1987)). The latest version of Meteorology-Chemistry Interface Processor (MCIP 4.3) was used to post-process WRF results and prepare input data for CMAQ (Otte and Pleim, 2010).The initial and boundary chemical conditions were derived from Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). Downscaling from MOZART to CMAQ was developed and discussed in detail in an upcoming paper (Ma et al., in preparation). The emissions of air pollutants in China were estimated by Tsinghua University (Wang et al., 2014;Zhao et al., 2013;Zhao et al., 2017;Zhao et al., 2018) using an "emission factor method". The provincial emissions from area and mobile sources were calculated from the activity data (energy consumption, industrial product yields, solvent use, etc.), technology-based uncontrolled emission factors, and penetrations of control technologies, and subsequently distributed to the model grids according to the spatial distribution of population, GDP, and road networks. A unit-based method is applied to estimate and locate the emissions from large point sources including power plants, iron and steel plants, and cement plants. The emissions from open burning of agricultural residue have been included in the anthropogenic emission inventory developed by Tsinghua University. They were calculated based on crop yields, the 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 from natural forest and grassland fires was ignored in this study, primarily due to the relatively small contribution in particular over the North China (Qin and Xie, 2011). Biogenic emissions were calculated by the

Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006). For each month of the CMAQ simulations, a week of model spin up was used to reduce the influence of the initial conditions.

3. Monthly variations of $PM_{2.5}$ concentration

3.1 Difference of $PM_{2.5}$ in December 2015 and January 2016

Fig. 1 shows the spatial distribution of $PM_{2.5}$ concentration in December 2015 (Fig. 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 2014-2016). The three-year average $PM_{2.5}$ concentration in December (2014-2016) and January (2015-2017) was shown in Fig. S1 in the supporting information. Consistent with previous studies (Chang et al., 2016;Yuan et al., 2017;Zhang et al., 2017), severe haze pollution occurred in December 2015 particularly over the North China Plain (NCP) (black box in Fig. 1a excluding Bohai area), with monthly mean $PM_{2.5}$ as high as 148 μg/m^3, much higher than that in December 2014 (97 μg/m^3) and slightly higher than that in December 2016 (135 μg/m^3). In January 2016, the anomaly (relative to January from 2015-2017) of $PM_{2.5}$ showed large decrease compared to the anomaly of December 2015 (relative to December from 2014-2016) over the NCP, with a mean decrease of 35 μg/m^3, whereas the $PM_{2.5}$ anomalies increase in January of 2015 (33 μg/m^3) and 2017 (3 μg/m^3) relative to the adjacent December anomaly (Fig. S~~1~~2 in the supporting information).

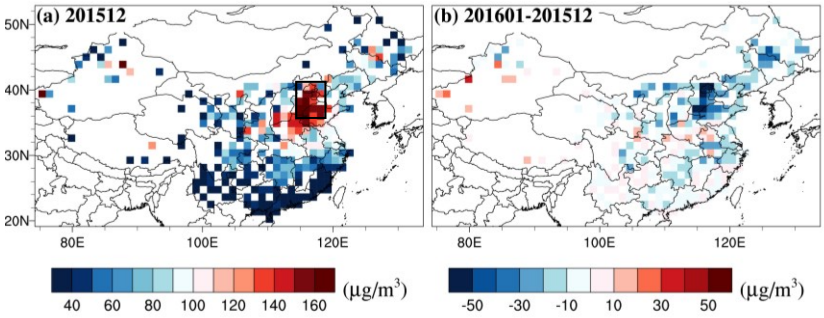

Fig. 1. Monthly mean $PM_{2.5}$ concentration in December 2015 (Fig. 1a), and the difference (Fig. 1b) between the anomaly of January 2016 (relative to January from 2015-2017) and December 2015 (relative to December from 2014-2016). The black box in Fig. 1a indicates the area of NCP.

3.2 $PM_{2.5}$ anomaly from long time series

To further test the robustness of the large drop of $PM_{2.5}$ from December 2015 to January 2016, monthly mean anomaly of $PM_{2.5}$ relative to 2009-2017 over U.S. Embassy Beijing is shown in Fig. 2. The $PM_{2.5}$ anomaly in December is generally negative (i.e., low $PM_{2.5}$ concentration) from 2009-2014 except for a substantial positive value of 48 μg/m^3 in 2015. For the January $PM_{2.5}$ anomaly, the large positive value in 2013 has been widely studied to investigate the mechanisms modulating the severe haze events (Han et al., 2016;Kajino et al., 2017;Shi et al., 2017;Ye et al., 2016). Looking at the difference between the January and December anomalies, the exceptional difference between January 2016 and December 2015 stands out, with the $PM_{2.5}$ concentration anomaly (-38 μg/m^3) in January 2016 showing a decrease of -86 μg/m^3 relative to the anomaly in December 2015 (48 μg/m^3), which is much more negative than the difference in any other years and far more negative than the 99% confidence interval value of -66 μg/m^3.

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 calculated relative to a longer period despite the short record of the AQI data. Similar to Fig. 2, abnormally high concentration in December 2015 and low concentration in January 2016 are also found in the API/AQI record (Fig. S3 in the supporting information). As anthropogenic emissions such as NO_x, SO_2, and primary $PM_{2.5}$ have been steadily decreasing since 2011 (Liu et al., 2016;Wang et al., 2017), the abnormally high $PM_{2.5}$ concentration in December 2015 requires an explanation. Moreover~~ interestingly~~, the anthropogenic emissions ~~of SO_2~~ in January could be comparable to or higher than that in ~~is usually higher than~~ December~~ primarily due to a higher power demand~~, i.e., in ~~and this is the case for~~ January 2016, ~~with a~~ higher SO_2 concentration, implicative of SO_2 emissions, was found than December 2015 based on observed data (http://www.pm25.in; not shown). Thus, what mechanism triggers the sharp decrease of haze in January 2016 compared to December 2015 needs to be investigated.

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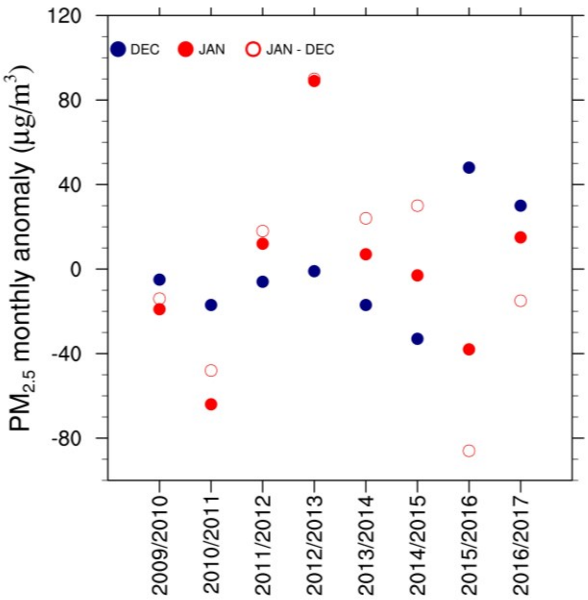

Fig. 2. Monthly mean anomaly of $PM_{2.5}$ in the adjacent December (solid blue) and January (solid red) as well as the change (hollow red circle) from December to

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 $PM_{2.5}$

4.1 The effect of meteorological modulation

To determine if meteorological factors play a role in the anomalous December to January change in $PM_{2.5}$, we first examined the mid-tropospheric circulation system during December 2015 and January 2016. As shown in Fig. 3a for December 2015, the northeastward shift of Siberian High and anticyclonic high pressure system in the NCP reduced the northerly wind transporting cold air from Siberia to the NCP, favoring the haze formation (Cai et al., 2017;Chang et al., 2016). In January 2016 (Fig. 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 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 with some level of predictability.

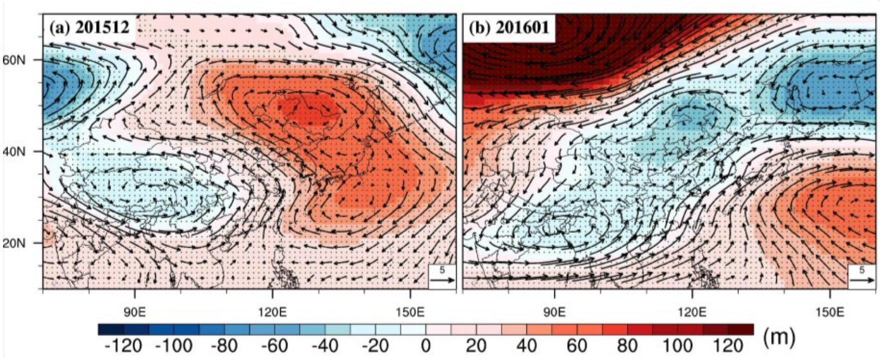

Fig. 3 Anomaly of geopotential height and wind vector at 500 hPa (unit: m/s) for December 2015 (Fig. 3a) and January 2016 (Fig. 3b), with~~. (The~~ anomaly~~ is~~ relative to the respective month in1987-2016~~)~~. Stippled areas indicate geopotential height

exceeding 90th 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). The black box in Fig. 3a indicates the area of NCP.

4.2 Possible linkage with climate variability

From the Nino 3.4 index, defined as the 3-month running regional mean SST anomaly over the tropical region (5 ºS-5 ºN, 170º-120ºW) relative to 1951-2000 (Rayner et al., 2003), the El Niño signal reached a peak in December 2015, and started to decay in January 2016. The development and decay of this El Niño event and the associated change in circulation pattern have been well documented by Xue and Kumar (2017). The 2015-2016 El Niño event is known as a super strong event comparable to the other two super events during 1982/1983 and 1997/1998 (Ren et al., 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 examined the super El Niño events in 1982/1983 and 1997/1998 based on Nino 3.4. In 1997/1998, the El Niño peaked in December 1997 and started to decay in January 1998, but in 1982/1983, the El Niño peaked in January 1983 and started to decay in February 1983. The 500 hPa geopotential height anomalies during these four months are shown in Fig. S~~3~~4. Consistent with the anomalous circulation features in Fig. 3, an anticyclonic circulation anomaly dominates over the NCP during the peak of the El 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 anomaly patterns as climatological features, we checked the 500 hPa anomalies in December, January, and February during the past thirty years (1987-2016) but we could not find similar opposite patterns in adjacent winter months during any other years. Moreover, the ensemble mean circulation anomaly during the peak and decay of the El Niño events from 1948-2016 was shown in Fig. S5 in the supporting information, and only the composite of three super El Nino events

(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 hints at the possible linkage between the anomalous circulation patterns and super El Niño.

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 surface air temperature during the adjacent months of the super El Niño events (January 1983, December 1997 and 2015 versus February 1983 and January 1998 and 2016) are shown in Fig. 4, while anomaly of near surface wind at 10-m is shown in Fig. S6 in the ~~5~~supporting information. These figures clearly depict opposite anomaly patterns in the NCP, showing southerly anomaly (Fig. 4~~5~~a; Fig. S6~~5~~a) during the peak of the El Niño, and northerly anomaly (Fig. 4~~5~~b, Fig. S6b) during the start of the decay phase. The southerly wind anomaly, abnormally warm near surface air temperature 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 plain and Siberia and reduced pressure contrast between the Asian continent and the 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. S6b) when El Niño starts to decay. The low-level circulation patterns and near surface air temperature are consistent with the seesaw changes of 500 hPa geopotential height 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 haze formation.

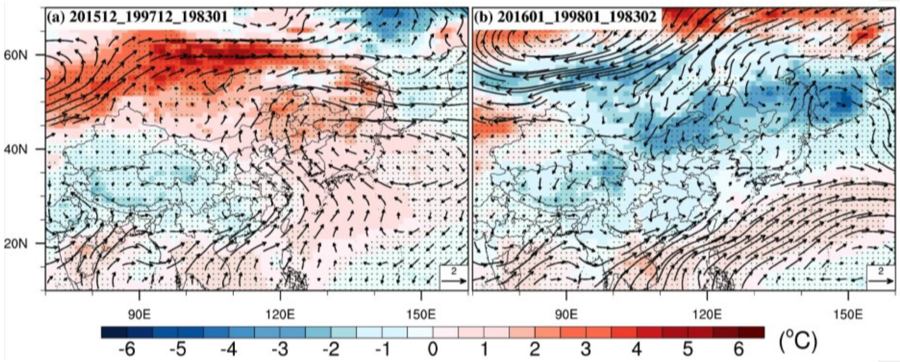

Fig. 4 850 hPa wind anomaly (unit: m/s) and 2-m air temperature anomaly (relative to 1987-2016). The anomaly was conducted over the respective month from 1987-2016, i.e., mean value of anomaly over December 2015, December 1997 and January 1983 (Fig. 4a), and January 2016, January 1998 and February 1983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

4.3 Sensitivities of $PM_{2.5}$ to meteorological conditions

As shown in the above analyses, the meteorological conditions in December 2015, December 1997, January 1983 exhibit a clear contrast with those of the following 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 time periods under the same emissions, with a spatial resolution of 36 km by 36 km. The simulated meteorological conditions including near surface (2-meter) air temperature (T2) and specific humidity (Q2), 10-m wind speed (WS10) and direction (WD10) were evaluated using the NCEP Meteorological Assimilation Data Ingest System (MADIS; https://madis.noaa.gov) data on hourly time scale. Using the benchmark based on Emery et al. (2001), the meteorological parameters compared 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 (19.25°) of wind direction at 10-m (WD10) in December 2015 is partly attributable to the model comparison with observed values close to 0° or 360°, which may yield large bias albeit the small differences in reality (i.e., 10° in model vs. 350° in observation).

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

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

* 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

MCIP was used to prepare input for CMAQ. A code bug in MCIP 4.3 for processing land use information was found that affected the percentage of urban area (PURB), leading to low $PM_{2.5}$ concentration in the simulations over urban areas (see the part 2 of the s~~S~~upporting i~~I~~nformation for more details). With the bug fix, the concentration of $PM_{2.5}$ matches the observations well (Fig. 5~~6~~). Statistical metrics such as mean fractional bias/error (MFB/MFE) were used to evaluate the simulation of $PM_{2.5}$, as recommended by US EPA (USEPA, 2007). Based on almost 200 observational sites in North China, the MFB/MFE of $PM_{2.5}$ from CMAQ is 1%/55% and 1%/56%, respectively for December 2015 and January 2016, satisfying the benchmark of 50%/75% (USEPA, 2007). More evaluation of $PM_{2.5}$ was discussed in part 3 of the supporting information.

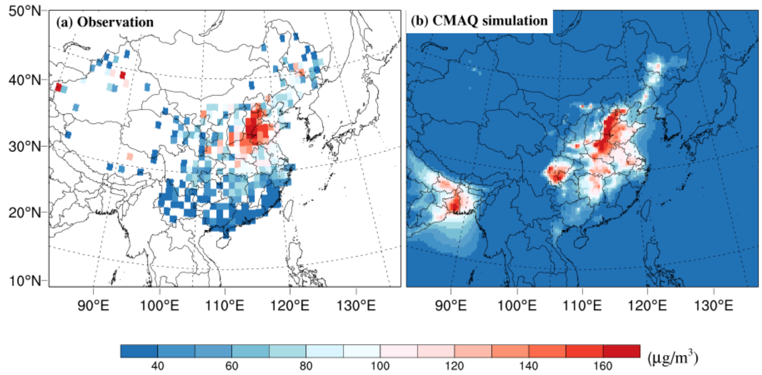

Fig. 5~~6~~. Spatial distributions of monthly mean $PM_{2.5}$ concentration in December 2015 for observations (a) and CMAQ simulation (b). Please note that the label bar starts from 40 μg/m^3 instead of 0 in order to make the comparison easily viewed.

Fig. 6~~7~~ shows the spatial distribution of monthly mean $PM_{2.5}$ in December 2015 (Fig. 6~~7~~a), December 1997 (Fig. 6~~7~~c), January 1983 (Fig. 6~~7~~e) and the differences compared to the $PM_{2.5}$ in the following month. Although the same emissions at the level of December 2015 were used across the simulations, the results clearly show much higher monthly mean $PM_{2.5}$ in December 2015 (Fig. 6~~7~~a), December 1997 (Fig. 6~~7~~c), January 1983 (Fig. 6~~7~~e) particularly over the NCP compared to the following month (Figs. 6~~7~~b,d,f), with mean reduction in the NCP of 30-50 μg/m^3 or more. In addition, by applying the process analysis (not shown), we found that the dry and wet deposition contributed a total of 12 % to the total removal processes over NCP in December 2015, comparable to that (13%) in January 2013, indicating the seesaw patterns was not modulated by the deposition. Hence the modeling results further verified the effect of the meteorological conditions on the seesaw $PM_{2.5}$ pattern.

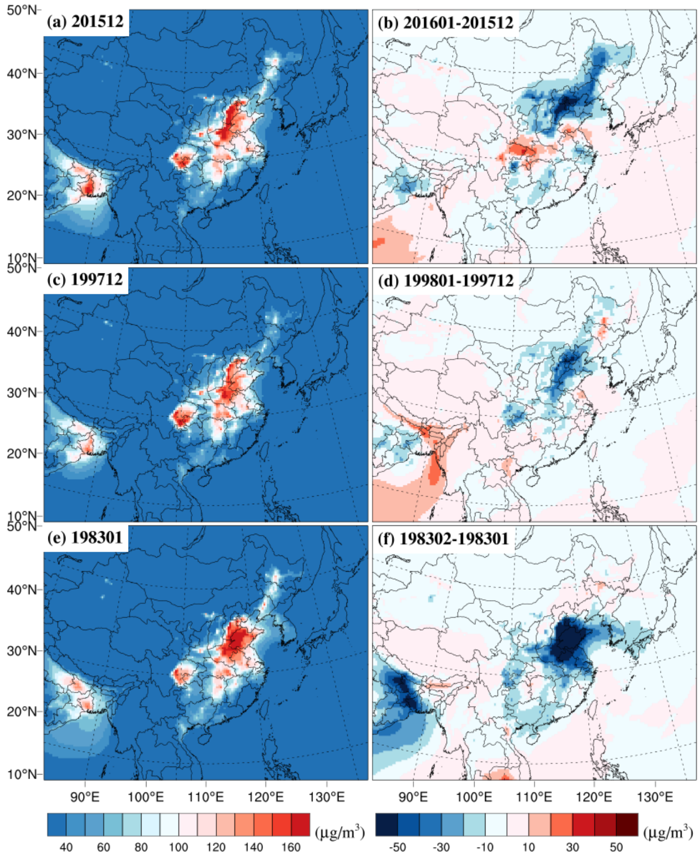

Fig. 6~~7~~ Monthly mean $PM_{2.5}$ during the three super El Niño events, i.e., December 2015 (a), December 1997 (c), January 1983 (e) and the differences of $PM_{2.5}$ compared to the following month (b, d, and f)

4.4 Synthesis of the mechanism modulating the $PM_{2.5}$ variations

The different modulation effects of the development and decay phase of El Niño 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 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 June and July, enhanced convection in August over the warm pool significantly reduces the 500 hPa geopotential height and pushes the WPSH to retreat eastward substantially. For intraseasonal variation in winter, Nie et al. (2016) found that the change from warm anomaly in December 2015 to cold anomaly in January 2016 in the NCP is possibly associated with the sudden shift of the Arctic Oscillation (AO) from a positive phase to a negative phase in January 2016, which is dynamically 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 strong during the negative phase of the AO, but a transition of the AO to a positive phase in January 2013 caused a sudden weakening of cold advection and increased stagnation. The resulting severe haze in January 2013 has been widely discussed previously (Cheng et al., 2017;Kajino et al., 2017).

The EAWM is closely related to winter haze conditions in the NCP. Cheung et al. (2012) found that in general, the EAWM is ~~enhanced (~~weakened~~)~~ (enhanced) when the AO and ENSO are in phase (out of phase). They also noted that a similar relationship at sub-seasonal time scale may be further investigated considering the possibility of a sharp reversal of the AO resulting from tropospheric–stratospheric interaction (Baldwin and Dunkerton, 1999). To delve into the mechanism modulating the sudden reversal of the AO from positive to negative phase during the decay of the mature phase of El Niño, anomaly zonal wind changes at 200 hPa are shown in Fig. 7~~8~~. Climatologically (i.e., 1987-2016), the subtropical jet is centered around 35-40ºN in December and shift southward in January based on ERA-Interim (not shown). A clear dipole feature of the sub-seasonal changes of anomaly zonal wind with negative change north of 40ºN indicates an obvious anomalous sub-seasonal southward shift of the subtropical jet during the three El Niño events from its climatological sub-seasonal shift. The subtropical jet shift has been confirmed by Geng et al. (2017) using climate model simulations, showing phase transition of AO from positive to 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 stratospheric polar vortex and the subsequent negative phase of the AO (Bell et al., 2009;Fletcher and Kushner, 2010), leading to more cold advection, enhanced EAWM, and higher PBL height compared to the conditions during the peak of the El Niño with lower PBL height (Fig. 8~~9~~a). These changes in the meteorological conditions associated with the changes in the El Niño development and AO phase shift and the consequent changes in $PM_{2.5}$ concentration are summarized schematically in Fig. 9~~10~~. In short, during the mature phase of El Niño (Fig 9a; indicated by the red Nino 3.4 index in the bottom of Fig. 9), the El Niño and positive AO (+AO) are in phase and weakens the EAWM (with orange and blue arrows indicating effect from El Nino and AO, respectively in Fig. 9a), leading to lower PBL and the subsequent severe haze pollution. After the mature phase of El Niño (Fig. 9b), AO suddenly turns to the negative phase (-AO), enhancing the EAWM (blue arrows in Fig. 9b) and subduing the weakening effect from El Nino (the southerly orange arrow) on EAWM, resulting in low $PM_{2.5}$ concentration.

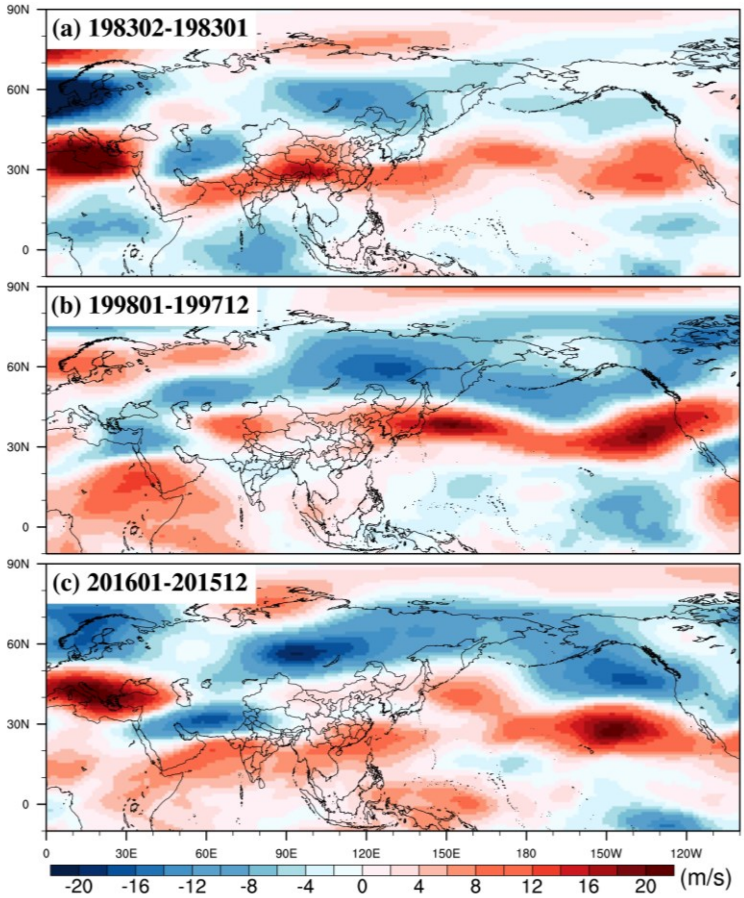

Fig. 7~~8~~ Sub-seasonal changes of anomaly of 200 hPa zonal wind (relative to the respective month in 1987-2016) during the start of the decay for three super El Niño events, including the differences between anomaly of February 1983 and January 1983 (Fig. 7a), January 1998 and December 1997 (Fig. 7b), January 2016 and December 2015 (Fig. 7c)

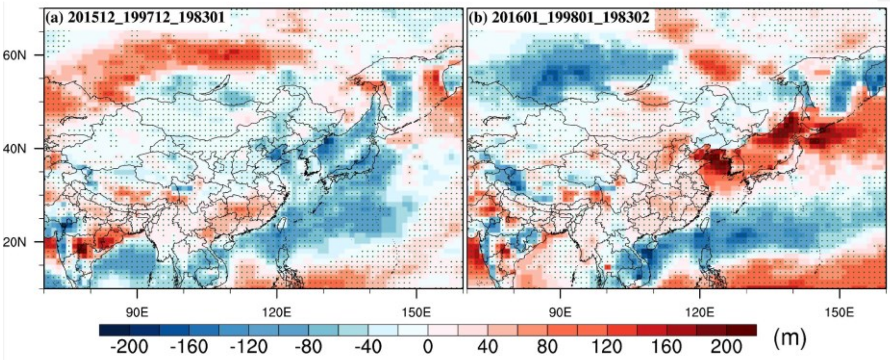

Fig. 8~~9~~ Sub-seasonal changes of PBL height anomaly (relative to 1987-2016). The anomaly was conducted over the respective month from 1987-2016, i.e., mean value of anomaly over December 2015, December 1997 and January 1983 (Fig. 8a), and January 2016, January 1998 and February 1983 (Fig. 8b). ~~(relative to 1987-2016);~~ S~~s~~tippled areas indicate exceedance of 90th confidence interval, with the same method used as Fig. 3.

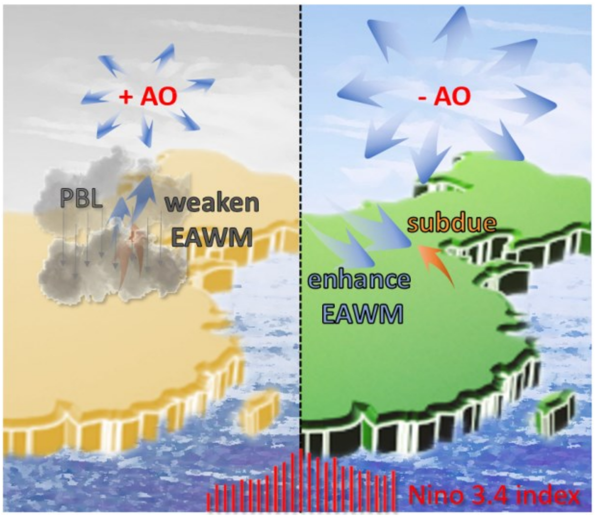

Fig. 9~~10~~ Schematic of the modulation of El Niño and Arctic Oscillation on atmospheric circulation and haze in northern China

Summary and discussions

This study identifies a distinct seesaw feature of abnormally high and low monthly mean $PM_{2.5}$ concentration in the two consecutive months of December 2015 and January 2016, respectively, in the North China Plain. Accompanying the seesaw features of $PM_{2.5}$ are opposite large scale circulation patterns with positive (negative) anomaly of 500 hPa geopotential height and southerly (northerly) wind anomalies at the low level (850 hPa) over the North China Plain during December 2015 (January 2016). The contrast in $PM_{2.5}$ between December 2015 and January 2016 is significantly larger than the change from December to January between 2009 and 2017 recorded at US embassy Beijing.

As the modulation of meteorological conditions on $PM_{2.5}$ occurred during a strong El Niño period, we explored the relationship between strong El Niño and $PM_{2.5}$. First, using a regional climate/chemistry model WRF/CMAQ, we identified that all three super El Niño events in the recent record (1982/83, 1997/98, and 2015/16) yield similar seesaw modulation features of $PM_{2.5}$. Further analysis showed that the seesaw $PM_{2.5}$ variations are modulated by the combined effect of El Niño and Arctic Oscillation (AO). In December 2015, the mature phase of an extreme El Niño, accompanied by a positive AO, weakened the EAWM, as indicated by a positive 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 in reduced planetary boundary layer (PBL) height, abnormally warm temperature and substantial haze accumulation during this period. In the following month (January 2016) when the El Niño began to decay, a sharp reversal of the AO from a positive phase (in December 2015) to a negative phase triggered enhanced EAWM, inducing cold advection and anomalous low-level northerly winds over the NCP that fostered 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 extreme El Niño events during the corresponding decay period (February 1983 and January 1998) after the mature phase (January 1983 and December 1997). These changes in circulation are likely associated with a southward shift of the upper troposphere jet at 200 hPa and weakened stratospheric vortex during the decay phase compared to the mature phase of El Niño. As the frequency of super El Niño like the 2015/16 event is projected to increase in the future (Cai et al., 2014;Cai et al., 2015), the seesaw modulation of super El Niño and AO may become more frequent, revealing vital information useful for policy makers dealing with air quality issues in China.

Although we performed WRF-CMAQ simulations to demonstrate the impacts of 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 limitation, future studies will design Atmospheric Model Intercomparison Project (AMIP) type scenarios, e.g., by running multi-ensemble members of scenarios with global models such as Community Earth System Model (CESM) using different SSTs 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 and AO on haze formation.

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

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