Response to Comments of Reviewer A

Manuscript number: acp-2019-1194

Author(s): Juan Feng, Jianlei Zhu, Jianping Li, and Hong Liao

Title: Aerosol concentrations variability over China: two distinct leading modes

Response to Reviewer A

Overview comment:

This paper studied the month-to-month variability of aerosol concentrations (AC) over China using a GESO-Chem model. The emission level in the model is set to a constant level of the year 2005. They found that two distinct lead modes dominate the natural variability: one is monopole mode which is related to the 3-month leading ENSO, while the second one is meridional dipole mode which is related to the NAO. The underlying physical mechanism is further analyzed. The results show that dynamical stability associated with the change of low-level convergence and planetary boundary layer height and thermal condition both play important roles.

Overall, the paper is well written and easy to understand. The topic is perfectly in line with ACP journal. Therefore, I recommend publishing after a minor revision.

Response to general comment:

Thanks to the reviewer for the helpful comments and suggestions. We have revised the manuscript seriously and carefully according to the reviewer's comments and suggestions. More details could be found in the revised manuscript.

Specific comment:

1. *Title: Since this paper focus on the internal climatic variability, the emission level is fixed at the year 2005. Otherwise, the first leading mode might show an increasing trend due to the dominate role of anthropogenic emissions according to the previous study. Therefore, I suggest a title changed to: Aerosol concentrations natural variability over China: two distinct leading modes.*

Response:

Thanks. We agree with the reviewer's comment that the natural variability of the aerosol concentrations is discussed in the present work, however, the aerosol concentrations is mainly the anthropogenic emissions. In addition, the natural aerosols, for example, mineral dust is not included in the manuscript. To avoid the misunderstanding, we have not changed the title.

2. *Line 247 and other places: "emission" is not approximate here. How about using "transmission"?*

Response:

This has been revised.

3. *Line 309: month-to-month variability of AC*

Response:

This has been revised.

Response to Comments of Reviewer B

Manuscript number: acp-2019-1194

Author(s): Juan Feng, Jianlei Zhu, Jianping Li, and Hong Liao

Title: Aerosol concentrations variability over China: two distinct leading modes

Response to Reviewer B

Overview comment:

This paper tries to identify the climatic contribution to monthly aerosol variability over China using the GEOS-Chem model and tele-connection methodology. Though generally well written, this reviewer finds that more work needs to be done before it can be published. The major concerns include 1) model evaluation – there is a limited meteorological evaluation (against NCEP reanalysis and Hadley SST) but no evaluation of aerosol simulation at all, which makes audiences hard to gauge how meaningful the result is; 2) emissions – the authors fixed the emissions at the 2005 level to single out the climatic effect. However, aerosol has profound effects on climate through aerosol-cloud-radiation interactions. Different level of aerosol should have different feedback in the climate system. Ideally, the authors should conduct two experiments with high and low emissions to draw the conclusion. If that is not possible, the authors should at least discuss this point in the manuscript; 3) a few statements in the text were not consistent with the corresponding figures; and 4) driving meteorology – it appears GEOS-4 meteorology has been utilized to drive GEOS-Chem, which is very outdated. Why was the GEOS-5 not used?

Response to general comment:

Thanks to the reviewer for the helpful comments and suggestions. We have revised the manuscript seriously and carefully according to the reviewer's comments and suggestions. We would like to clarify the above comments in the following points,

1. As to the comment "*model evaluation – there is a limited meteorological evaluation (against NCEP reanalysis and Hadley SST) but no evaluation of aerosol simulation at all, which makes audiences hard to gauge how meaningful the result is*"

1) **As to the reliability of the meteorology fields**: we have shown in the manuscript, the input surface skin temperature of GEOS-Chem is highly correlated with the widely used SST dataset HadISST, in both spatial distribution and the long-term variability. Moreover, the input meteorological fields, such as winds, temperature, humidity, have been evaluated in Zhu et al. (2012a, 2012b) and Feng et al. (2016, 2017, 2019), and the result suggested the GEOS-Chem input meteorological fields are highly consistent with the NCEP/NCAR reanalyses. The above result provides confidence for the reliability of the meteorological fields of GEOS-Chem model.

2) **As to the reliability of the model simulations**:

a) Because of lacking valid aerosol concentrations observational data, we have not shown the comparison between the observations and simulations. However, previous studies have reported that the GEOS-Chem could well capture the seasonal and interannual variations of aerosol concentrations and O_3 over eastern China (including southern China; e.g., table 1 & fig. 1 in *Zhang et al.*, 2010; fig. 2 in *Wang et al.*, 2011; table 4 in *Lou et al.*, 2014; fig. 2 in *Yang et al.*, 2014)). In this study, we focused on the relative influences of climatic event on the aerosol concentrations instead of the absolute values of aerosol concentrations.

b) We have adopted the reviewer's comment by further comparing the spatial distribution of the aerosol concentrations based on the model simulations and MODIS AOD data (Figure R1). It is seen that similar spatial distributions are observed, e.g., the maximum is located to the north of Yangtze River during January, and to the north of Huanghe during July. That is both the seasonal evolution and spatial distribution of the aerosol concentrations over China is well reproduced in the GEOS-Chem.

c) We have compared the absolute values of aerosol species (i.e., Nitrate) based on the simulations and observations from other works (Table R1). It is seen that the simulated values of Nitrate in China based on the simulations and observations are equivalent.

The above discussions provide confidence for employing the GEOS-Chem to explore the influences of climatic events on aerosol concentrations, and it is proved to be a useful tool to understand the impacts of climatic event on aerosol concentrations without enough observations.

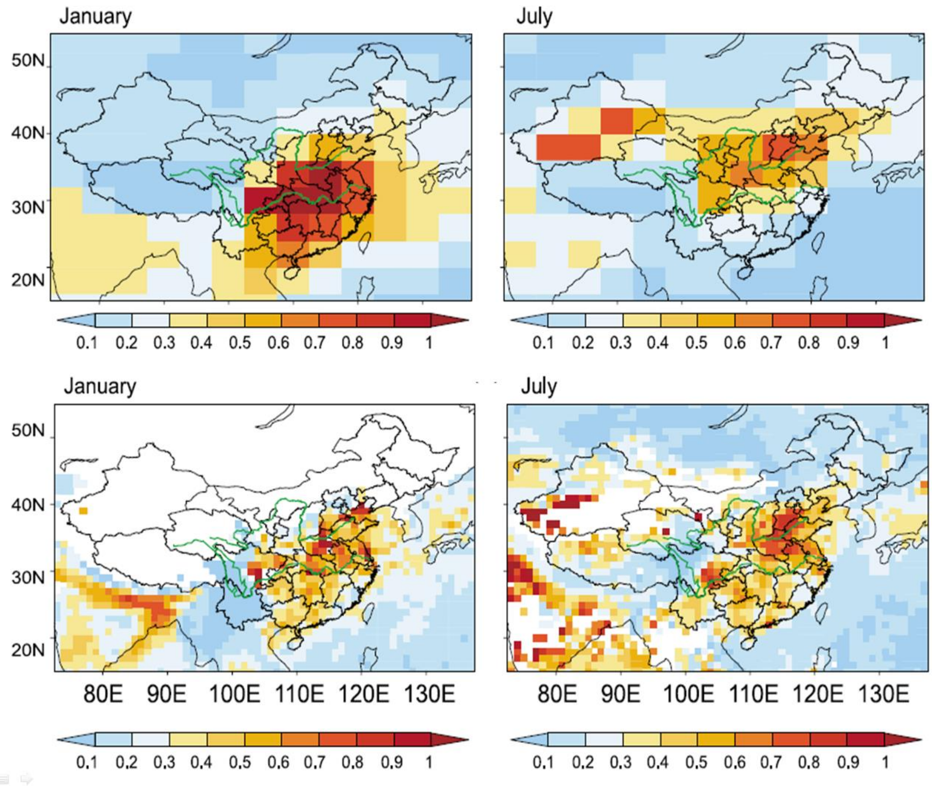

Figure R1. (Upper) The simulated aerosol concentrations over China during (left) January in 2001 and (right) July in 2001. (Below) As in the upper, but based on the MODIS AOD.

Table R1. The Nitrate concentrations based on the simulations and observations.

Location	**Value** (μg m^{-3})				**Reference**
	Summer		Winter		
	Simulation	Observed	Simulation	Observed	
Beijing (121.3°E,31.1°N)	9.82	11.18±10.37	10.71	12.29±12.12	Wang et al.,2007
Shanghai (121.3°E,31.1°N)	7.07	4.76	17.95	10.10	Ye et al., 2003
Xi'an (108.6°E,34.2°N)	3.43	14	13.23	56	Zhang et al., 2002
Nanjing (118.5°E, 32°N)	12.64	3.24	37.22	12.9	Yang et al., 2005
Fuzhou (119.3°E,26.1°N)	1.35	1.10±0.35	10.64	8.77±3.17	Xu et al. 2011
Hong Kong (114°E, 22.5°N)	0.89	0.85±0.60	6.48	2.65±2.33	Louie et al., 2005

2. As to the comment "*emissions …… Ideally, the authors should conduct two experiments with high and low emissions to draw the conclusion. If that is not possible, the authors should at least discuss this point in the manuscript*",

Thanks for the comment. The reviewer is right that aerosol has profound effects on climate through aerosol-cloud-radiation interactions, we have adopted the reviewer's comment by fixing the emissions at the level of year 1986 (low emission situation) and year 2006 (equivalent level of year 2005) to compare the role of different emissions on the distribution of aerosol concentrations. Four experiments are designed, meteorology field fixed on 1986 (M86), emission level at 1986 (Inv86) and at 2006 (Inv06), and meteorology field fixed on 2006 (M06), emission level at 1986 (Inv86) and at 2006 (Inv06). It is seen that aerosol concentrations significantly increased when the emission level increased, and the aerosol concentrations show a similar increasing trend under different meteorological conditions when the emission increases (Table R2). Note that the annual SO_2 emissions in eastern China increased from 4.54 Tg S yr^{-1} in 1986 to 12.01 Tg S yr^{-1} in 2006, with an increase rate of 164.5%, which is paralleling to the increase rate of SO_4^{2-}. Similar increase is seen in other particles.

On the other hand, it is seen that although the emission level is same, e.g., Inv06 but in M86 and M06, Inv86 but in M86 and M06, the output aerosol concentrations are different, suggesting that the role of meteorological conditions in impacting the aerosol concentrations. This point confirms the important modulation of the meteorological factors on the distribution of aerosol concentrations.

The above discussions indicate the role of meteorological conditions plays important role in impacting the distribution of aerosol concentrations. However, when the anthropogenic emissions have times increased, the variation of aerosol concentrations is mainly attributed to the emissions.

We have included the discussions into the revised manuscript for a better presentation.

Table R2. The differences of aerosol concentrations under different anthropocentric emissions averaged over eastern China (110°–125°E, 20°–45°N) during boreal winter.

EX	M86					M06			
	concentrations ($\mu g\ m^{-3}$)			Percentage		concentrations ($\mu g\ m^{-3}$)			Percentage
species	Inv06	Inv86	Difference	(%)		Inv06	Inv86	Difference	(%)
SO_4^{2-}	6.42	3.19	3.23	101.13		7.07	3.63	3.44	94.75
NO_3^-	13.88	5.90	7.98	135.35		15.43	6.74	8.69	128.87
NH_4^+	6.40	2.90	3.50	120.85		7.09	3.3	3.79	114.9
BC	1.16	1.16	<0.01	<0.02		1.24	1.24	<0.01	<0.01
OC	1.71	1.70	<0.01	<0.30		1.8	1.8	<0.01	<0.13
$PM_{2.5}$	29.56	14.84	14.72	99.19		32.64	16.71	15.92	95.28

3. As to the comment "a few statements in the text were not consistent with the corresponding figures",

We carefully checked the manuscript and all the figure captions in the revised manuscript.

4. As to the comment *"driving meteorology – it appears GEOS-4 meteorology has been utilized to drive GEOS-Chem, which is very outdated. Why was the GEOS-5 not used?",*

We agree with the reviewer that the GEOS-4 is an old version of the model. The GEOS-5 is a relative new version; however, it is only available for the period 2004-2013. We have explored the month-to-month variability of the aerosol concentrations in this study, ten years data is not enough for a climatological analysis. Moreover, as mentioned above, the GEOS-4 is highly consistent with the widely used reanalysis dataset, which provide the reliability of the present work.

Major comment:

1. *Lines 116 – 124: More details are needed for model description, e.g., driving meteorology, emissions inventories, etc. Were the biogenic emissions included in the simulation? If so, was it online calculated? Meteorology plays an important role in regulating biogenic emissions. A 2 x 2.5 degree model resolution appears very coarse for aerosol simulation. Were the results expected to change if a higher resolution simulation were conducted and analyzed?*

Response:

Thanks for the comment. The biogenic emissions are included in the simulation and it is online calculated. As to the resolution of the model simulations, as mentioned this model is widely employed to explore the aerosol concentrations variations all over the world, including East Asia, and it is proved to be a useful and reliable tool to investigate the variation of the aerosol concentrations. Due to the compute capability and time limitation, we have not updated the resolution of the simulation, however, we will adopt the reviewer's comment and expect to update the simulation to a higher resolution to further examine the influence of meteorological conditions on the aerosol concentrations.

2. *Line 137: mineral dust is an important contributor to aerosol loading at least in spring over the northern China. Would exclusion of dust skew the results?*

Response:

The reviewer is right, mineral dust is an important contributor to the boreal spring aerosol concentrations over northern China. However, the month-to-month variability is explored in this study, rather than the spring. That is the time scale are different. In addition, mineral dust is mainly a kind of natural aerosols, however, we focused mainly the anthropogenic aerosols in this study.

3. *It is not clear why the authors correlated PC1 to Nino 3.4 index while correlated PC2 to NAOI. The explanation in lines 185 ~ 197 was not convincing. Have the alternative correlations been tried?*

Response:

We have performed the correlations between the PCs and other climatic indices, e.g., Indian Ocean Dipole index, ENSO Modoki index, NAO index, and Niño3.4 index. Significant relationships are observed between the PC1 and Niño3.4 index, and between the PC2 and NAO index. The correlation coefficients between the PC1 and NAO index is 0.09, and it is 0.05 between the 3-month leaded Niño3.4 index and PC2, both are insignificant. Therefore, only the relationships between the NAOI and PC2, and between the Niño3.4 index and PC1 are discussed in the manuscript.

4. *Figure 3. Is the y-axis correlation coefficient? Add this information on the plots.*

Response:

We have adopted the reviewer's comment and replotted the figure.

5. *In Lines 226-227, add something like as shown in Figure 6 to reference that figure.*

Response:

This has been revised.

6. *Lines 238-242: Does the positive correlation of 3-month leading Nino 3.4 index and 700 hPa divergence mean anomalous convergence circulation?*

Response:

Figure 7 shows the spatial distribution of the correlation coefficients between the Niño3.4 index and convergence for the Niño3.4 index leading for 3 months. The positive correlations over the tropical eastern Pacific and negative correlations over western Pacific are corresponding to anomalous Walker circulation.

7. *Lines 246 – 247 and 270 (and in Conclusion): Should it be "not favorable for the emission of aerosol"? Are there any reference(s) for this statement?*

Response:

This has been revised, we have included the relevant reference.

8. *Line 250: should it be Figure 9a?*

Response:

This has been revised

9. *Line 260: should it be Figure 8?*

Response:

This has been revised.

10. *Line 275: should it be Figure 9b? (please make figure number consistent in the text and figure section).*

Response:

Thanks, we have carefully examined the figure captions and figure numbers in the manuscript during the revision.

11. *Line 278: It appears that the positive PBLH anomalies are not significant based on Figure 9b.*

Response:

The two figures present different physical variables, i.e., Figure 9 is for the PBLH, and Figure 10 is for the temperature. The two figures both for the regression distribution, but based on different variables. The statistical significance of the regression values was evaluated by a two-sided Student's t-test.

12. Lines 296 – 298: Figure 10b does not support this statement.

Response:

Figure 10b shows the relationship between the NAOI and temperature. It is seen that positive phase corresponds to opposite temperature anomalies over southern and northern China. Southern China is mainly occupied with positive anomalies, and northern China is under influenced by negative anomalies in general. This result agrees with the relation between the NAO and AC. Moreover, we have carefully examined the whole manuscript to avoid clerical errors during the revision process. The detailed revisions are shown in the revised manuscript. We hope the revised manuscript could offset the shortcomings in the original manuscript.

Aerosol concentrations variability over China: two distinct leading modes

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Abstract

Understanding the variability in aerosol concentrations (AC) over China is a scientific challenge and is of practical importance. The present study explored the month-to-month variability in AC over China based on simulations of an atmospheric chemical transport model with a fixed emissions level. The month-to-month variability in AC over China is dominated by two principal modes: the first leading mono-pole mode and the second meridional dipole mode. The mono-pole mode mainly indicates enhanced AC over eastern China, and the dipole mode displays a south-north out-of-phase pattern. The two leading modes are associated with different climatic systems. The mono-pole mode relates to the 3-month leading El Niño-South Oscillation (ENSO), while the dipole mode connects with the simultaneous variation in the North Atlantic Oscillation (NAO) or the Northern Hemisphere Annular Mode (NAM). The associated anomalous dynamic and thermal impacts of the two climatic variabilities are examined to explain their contributions to the formation of the two modes. For the mono-pole mode, the preceding ENSO is associated with anomalous convergence, decreased planetary boundary layer height (PBLH), and negative temperature anomalies over eastern China, which are unfavorable for emissions. For the dipole mode, the positive NAO is accompanied by opposite anomalies in the convergence, PBLH, and temperature over southern and northern China, paralleling the spatial formation of the mode. This result suggests that the variations originating from the tropical Pacific and extratropical atmospheric systems contribute to the dominant variabilities of AC over China.

1. Introduction

Aerosol particles are the primary pollutants in the atmosphere and play significant roles in influencing human health, environmental pollution, and regional and global climate (IPCC, 2013). The variation in aerosols shows considerable impacts on the climate via its direct and indirect effects by altering the radiation forcing and microphysical effects (e.g., Thompson, 1995; Zhang et al., 2011; Huang et al., 2006), indicating the important influences on the regional and global climate. For instance, it is noted that the 'cooling pool' in eastern-central China during the period 1960-1990 is partially attributed to increased aerosol concentrations (AC; Li et al., 2016), and that aerosols may exert influences on precipitation changes in both global and regional scales, as well as on monsoon systems (Rosenfeld et al., 2007; Cowan and Cai, 2011; Huang et al., 2014; Jiang et al., 2016; Lou et al., 2018). Thus, a better understanding of the AC variation is of significance for both scientific and practical efforts.

Meanwhile, the distribution and accumulation of aerosols are sensitive to meteorological conditions. The variations in the meteorological factors, e.g., precipitation, wind, temperature, planetary boundary layer height (PBLH), atmospheric stability, and humidity, could impact the AC by modulating the aerosol transport, deposition, and dilution processes (Aw and Kleeman, 2003; Lin and McElroy, 2010; Liao et al., 2015; Yang et al., 2017). The anomalies in meteorological conditions are attributed to the synoptic weather and climate systems. For the synoptic weather scale, Guo et al. (2014) indicated that stagnate weather conditions contribute to the periodic cycle of particulate matter events during boreal winter in Beijing. And the increase in relative humidity (Han et al., 2014) and decrease in the PBLH (Quan et al., 2014; Yang et al., 2015) would lead to an increase in the aerosols, thus contributing to the haze events during winter 2012 in northern China.

~~Meanwhile, the~~ The variations in the large-scale climatic systems, such as Pacific Decadal Oscillation (PDO), El Niño-South Oscillation (ENSO), East Asian summer and winter monsoon (EASM & EAWM), and North Atlantic Oscillation (NAO) show considerable effects in impacting the regional AC in both the seasonal and the interannual timescales. For example, researchers found that low values of AC are observed in Taiwan accompanied with the onset of the EASM (Chen and Yang, 2008). During the mature phase of the moderate La Niña event 2000/01, an anomalous south-negative-north-positive AC dipole pattern is seen over eastern China (Feng et al., 2017). The interannual variations in the EASM exhibit significant effects in impacting the summertime AC over China, i.e., high-level AC would be observed over eastern China along with a weaker EASM (Zhu et al., 2012; Lou et al., 2016; Mao et al., 2017). A similar situation is observed between the EAWM and AC over eastern China but during boreal winter, showing that a weaker EAWM relates to a high level of AC over China (Jeong et al., 2017). Zhao et al. (2016) have indicated that the decadal regime shift of the PDO showed significant role in impacting the decadal variations of boreal winter aerosols over eastern China. Feng et al. (2019) have reported the important influences of simultaneous ENSO and preceding autumn NAO signals on the winter AC over China by case study.

The above discussions highlight the effect of climate background in impacting the AC over China across different seasons, including signals from both the tropical and the extratropical, and originating from both the atmosphere and the ocean. However, the relative roles of climate systems are still unknown because there are strong interactions among the systems. For example, during the decaying summer of a warm ENSO event, a weaker EASM is expected to be observed (Wu et al., 2002), and the occurrence of a cold ENSO event during its mature phase is favorable for a stronger EAWM (Wang et al., 2008). The preceding spring (March to April) NAO indicates significant impacts on the following summer EASM in the interannual timescale (Wu et al., 2009). Moreover, the signals originating from the atmosphere (e.g., NAO, EASM, EAWM) and ocean (e.g., ENSO, PDO) present strong seasonality, prevailing in different seasons. As shown by the fact that AC over China are impacted by various climate systems, the relative importance of individual signals on their possible impacts in modulating the variability of AC remains unknown. ~~Meanwhile~~In addition, most of the previous studies regarding the influence of climate systems on AC focused on a certain season with little attention paid to spatial-temporal variability. These questions are important for improving the recognition of the modulation of climate systems on AC.

Consequently, one of the crucial motivations of the current work is to investigate the spatial-temporal variability in the monthly AC over China, highlighting the potential effects of climatic variabilities in modulating the spatial and temporal variations in AC, and understanding the possible physical processes involved. The rest of the study is arranged as follows. The model, datasets, and methods are presented in Section 2; The properties of the leading modes of AC variability are described in Section 3; Section 4 discusses the contribution of climatic modes on aerosol variabilities; and Section 5 provides the conclusions and discussions.

2. Datasets, model, and methodology

2.1 Model

The GEOS-Chem model is employed to detect the variability in AC over China. This model is a 3-dimensional tropospheric chemistry model with a 2.5° longitude × 2° latitude horizontal resolution and 30 vertical levels. The model is widely applied to investigate the potential modulation of climatic variabilities on the anomalous distributions of pollutants on various timescales, for example, on the seasonal (Generoso et al., 2008; Jeong et al., 2011; Feng et al., 2016, 2019), interannual (Jeong et al., 2017; Li et al., 2019), and interdecadal (Zhu et al., 2012) timescales. The high consistency in both the temporal and spatial distributions between the simulations and observations provides confidence for the feasibility of the present study.

As reported, the significant upward trend in anthropogenic emissions over China accounts for a large variance in pollutants, and the first dominant mode of boreal winter aerosols over eastern China represents anthropogenic emissions (Zhao et al., 2016). To highlight the modulation of the climatic variabilities on the variation in the aerosols, the anthropogenic and biomass burning emissions have been fixed at the year 2005 level. Thus, the variations in the aerosols in this context are attributed to the internal climatic variability.

The definition of particulate matter smaller than 2.5 μm in diameter (PM2.5) is followed by Liao et al. (2007),

$$[PM_{2.5}] = 1.29 \times [NO_3^-] + 1.37 \times [SO_4^{2-}] + [SOA] + [POA] + [BC]$$

where NO_3^-, SO_4^{2-}, SOA, POA, and BC are the aerosol particles of nitrate, sulfate, secondary organic aerosol, primary organic aerosol, and black carbon, respectively. Mineral dust and sea salt are excluded because these species are not the major components over China.

2.2 Datasets and methodology

The input meteorological variables of the model highly agree with the widely used atmospheric and oceanic datasets, i.e., the National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis (Kalnay et al., 1996), and the UK Meteorological Office Hadley Centre's sea ice and sea surface temperature (SST) datasets (HadISST; Rayner et al., 2003). These two datasets are employed to verify the climatic indices calculated based on the model input meteorological datasets. ENSO was characterized by the Niño 3.4 index, which is defined as the areal averaged SST over 120°W-170°W, 5°N-5°S. The monthly Niño 3.4 indices based on the HadISST and model input data are highly related with each other with a correlation coefficient of 0.99, confirming the reliability of the model data. The North Atlantic Oscillation index (NAOI) and Northern Hemisphere Annular Mode index (NAMI) are used to present the sea level pressure (SLP) oscillation between the mid-latitudes and high latitudes in the extratropical Northern Hemisphere. Following Li and Wang (2003), the NAMI is defined as the difference in the normalized global zonal-mean SLP between 35°N and 65°N, in which the 35°N and 65°N refer to the mid-latitude and high latitude, respectively. The definition of the NAOI resembles that of the NAMI but within the North Atlantic sector from 80°W to 30°E. Because the NAOI and NAMI are highly correlated with each other in both spatial distribution and temporal variation (Thompson and Wallace, 1998; Gong et al., 2001), the NAOI is utilized in the current context; similar results are obtained based on the NAMI.

Empirical orthogonal function (EOF) analysis was employed to obtain the spatiotemporal variability in monthly $PM_{2.5}$ over China. Correlation and regression are used to display the linkages between the variability in the $PM_{2.5}$ and the climatic modes. Here, the period 1986–2006 was taken as the climatological mean, and the annual cycle was removed before the analyses. The statistical significance of the correlation and regression values was evaluated by a two-sided Student's *t*-test.

3. Distinct leading modes of the variability in aerosol concentrations

3.1 Two leading modes

Figure 1 presents the spatial distribution of the first (EOF1) and second (EOF2) leading modes based on the monthly surface layer and column AC anomalies. A similar spatial distribution is observed in both the surface and column AC. The EOF1 and EOF2 modes explain 31.4% (37.0%) and 16.3% (14.1%) of the total variances for the surface layer (column) AC, respectively. Based on the *North*'s rule, the two dominant modes could be significantly separated from each other and from the rest of the eigenvectors based on the analysis of the eigenvalues in the light of sampling error above the 0.05 significance level. The rest of the modes are not discussed for their relative less explained variance or could not be well separated. The EOF1 mode displays a mono-sign pattern, with the maximum located in central eastern China (Figs. 1a and c). The EOF2 mode presents a meridional dipole pattern in eastern China, with opposite values to the south (positive values) and north (negative values) of the Yangtze River.

The temporal behavior of the two modes, the first and second principal components, i.e., PC1 and PC2, is displayed in Figure 2. Both PC1 and PC2 show strong interannual variations. The PCs based on the surface and column concentrations are closely correlated with each other, with coefficients of 0.80 and 0.79 for PC1 and PC2, respectively. The high consistency between the surface and column concentrations in both the spatial and temporal distributions implies that the factors governing their variations are the same. The maximum value of PC1 occurs in 1998, corresponding to the strongest El Niño event (1997/98) in the 20th century. For PC2, negative values are observed during the winters of 1989 and 2002, and positive values are observed during the winters of 1995 and 1997. However, the winters of 1989 and 2002 correspond to the positive polarities of the NAM or NAO, and the winters of 1995 and 1997 are paralleling to the negative polarities of the NAM or NAO. The potential linkage between the PCs and climatic variabilities is therefore analyzed. Here, the Niño 3.4 index is utilized to depict the variation of ENSO, and the NAOI (NAMI) is employed to reflect the variability in the NAO (NAM). Note that the indices based on the model input data are highly correlated with the observation datasets, and the monthly NAOI is closely related with the NAMI, exhibiting a significant correlation coefficient of 0.71 during period 1986-2006. Therefore, the NAOI is employed to detect the linkage between the PC2 and climate variability.

3.2 Linkage with the climate variabilities

Figure 3 displays the lead-lag correlation between the PC1 and Niño 3.4 index, and between the PC2 and NAOI to identify the linkage between the climatic variabilities and the two leading AC patterns. PC1 is significantly connected with the Niño 3.4 index, with the maximum occurring when the Niño 3.4 index is 3 months leading, implying a leading influence on PC1. The leading impacts of Niño 3.4 on the variation in PC1 are further seen from the seasonal evolution of the standard deviation in the corresponding indices (Fig. 4). The standard deviation of the monthly Niño 3.4 index shows that the maximum occurs during December, while the maximum occurs in March for that of PC1. The leading influences of Niño 3.4 on PC1 are further verified by the spatial distribution of correlations between PC1 and SST, as shown in Figure 5. For the correlation with the PC1 lagged for 3 months, significant positive correlations are observed over the tropical eastern Pacific and Indian Oceans, and negative correlations over the tropical western Pacific. The correlation pattern is like a canonical El Niño pattern. Note that the significant positive correlations over the tropical eastern Pacific gradually decrease as the SST leading time is reduced; however, the correlations over the tropical Indian Ocean become stronger, implying the effects of the Indian

Ocean capacitor along with the development of an ENSO event (Xie et al., 2009). The above result ascertains the preceding influence of ENSO on the variation in PC1, indicating a 3-month leading impact of ENSO on the following AC over China.

Meanwhile, the maximum negative correlation between PC2 and the NAO is simultaneous (Fig. 3b), implying a simultaneous impact of the NAO on the AC over China. Similar result is seen in the correlation between the NAMI and PC2. The simultaneous relationship between the PC2 and NAO is further estimated in their corresponding seasonal variation in the standard deviation (Figs. 4b and d). The maximum standard deviations of the NAO and PC2 both occur during January-February-March. A similar result is obtained based on the NAMI, suggesting significant negative impacts of the extratropical atmosphere variation on the AC over eastern China. Moreover, the correlations between the simultaneous PC2 and SLP display a negative NAO-like (NAM-like) structure, (Fig. 6), with significant positive correlations over the polar regions and negative correlations over the mid-latitudes. Note that this anomalous pattern is consistently observed in PC2s based on both the surface layer (Fig. 6a) and the column concentrations. (Fig. 6b).

The result above suggests that the variability in AC can be measured by climatic variabilities, of which the variation in EOF1 is linked to the 3-month leading SST variation over the tropical eastern Pacific, and that of EOF2 is related to the extratropical atmospheric variability-NAO. The possible physical process involved in their relationship is discussed in the following section.

4. Physical processes impacting on the leading modes

5.1 Circulation anomalies associated with ENSO

Figure 7 shows the anomalous circulations associated with ENSO to identify the atmospheric circulation process impacting the EOF1 patterns with the Niño 3.4 index leading for 3 months. It is seen that tropical eastern Pacific and southern China are controlled by significant positive correlations in the correlation with the convergence in the lower troposphere. That is, southern China and tropical eastern Pacific are influenced by anomalous convergence circulation under the influence of a 3-month leading ENSO signal. Meanwhile, tropical western Pacific is impacted by significant negative correlations, indicating that these regions are impacted by anomalous divergence. The anomalous convergence circulation over southern China is not favorable for the ~~emission~~transmission of AC. That is the anomalous circulation associated with 3-month leading ENSO signal would connect with enhanced AC over eastern China~~,~~ (Zhu et al., 2012; Feng et al., 2019), which agrees with the spatial distribution of EOF1. Moreover, the impacts of ENSO on the circulation is further seen in impacting the PBLH (Figure 8a). Significant negative ~~correlations~~anomalies are found over eastern China, indicating that the occurrence of a warm ENSO event would decrease the height of PBLH. The decreased PBLH relates to enhanced AC over eastern China. The above result suggests that the leading ENSO signal exhibits a significant role in affecting the circulation anomalies over China. Under the influence of warm ENSO events, the followed anomalous convergence and decreased PBLH over eastern

China are both unfavorable for the emission of AC, contributing to the formation of the EOF1 pattern.

5.2 Circulation anomalies associated with NAO

The anomalous divergence accompanied by the simultaneous NAO is presented in Figure 9. The northern Atlantic Ocean is influenced by an anomalous ~~tripole~~tri-pole structure, showing convergence-divergence-convergence anomalies from the polar region to the tropical regions. The occurrence of the anomalous circulation structure in the northern Atlantic Ocean is due to the fact that the variation in NAO would induce an anomalous ~~tripole~~tri-pole SST pattern within the northern Atlantic Ocean (e.g., Wu et al., 2009; Zheng et al., 2016) by which a downstream wave-train is expected to be observed (Ruan et al., 2015; Li and Ruan, 2018). The downstream wave train is seen with significant positive anomalies over southern China in the regression of NAOI to the divergence, while negative anomalies occur over northern China. That is, a positive NAO is accompanied with anomalous divergence (convergence) over southern (northern) China. The anomalous convergence over northern China is unfavorable for the emission of AC, corresponding to enhanced AC. However, the opposite situation is observed over southern China. The anomalous circulation connected with NAO further estimates the negative impacts of NAO on the EOF2 mode.

In addition, the potential impacts of NAO on PBLH over China are further examined. Figure 8b shows the anomalous PBLH regressed with reference to the NAOI to identify the role of NAO in determining the EOF2 mode. For a positive NAO phase, negative PBLH anomalies occupy northern China, suggesting a favorable condition for enhanced AC. In contrast, southern China is controlled by positive PBLH anomalies, paralleling the situation for decreased AC. The circulation anomalies connected with NAO in both the divergence and PBLH suggest that the impacts of NAO on the AC over northern and southern China are opposite, consistent with the spatial distribution of the EOF2 mode.

5.3 Role of temperature

Meanwhile, it has been reported that temperature shows an effect in impacting the distribution of aerosols. For example, it is reported that an increase in temperature is associated with a decrease in $PM_{2.5}$ over southern California (Aw and Kleeman, 2003) because enhanced temperature lead to decreases in organics and nitrate (Dawson et al., 2007). Accordingly, the associated impacts of the ENSO and NAO on the temperature over China are detected. Figure 10 displays the anomalous temperature regressed against the 3 months preceding Niño 3.4 index and simultaneous NAOI to detect the temperature anomalies connected with the two climate systems. For a warm event of ENSO, large areas of negative temperature anomalies occupy eastern China, with the maximum lying within the Yellow River and Yangtze River (Fig. 10a). The negative temperature anomalies imply a lower temperature condition, which would induce to enhanced AC.

For the NAO, its positive phase corresponds to opposite temperature anomalies over southern and northern China, being positive (negative) over the southern (northern)

China (Fig. 10b). Positive temperature anomalies over southern China parallels to a warmer situations and reduced AC in this region. Negative temperature anomalies over northern China set up a background of colder situations, which would increase the AC. The anomalous variation in the temperature agrees with the negative impact of NAO on the AC over eastern China. In addition, the temperature anomalies accompanied with the preceding ENSO are greater than those associated with the simultaneous NAO, highlighting the dominant role of ENSO in impacting the AC over eastern China.

5. Conclusions and Discussions

China has a high loading of aerosols and understanding the variability in AC is important not only for recognizing the interactions between aerosols and climate but also for scientifically understanding the current pollutant status. In the present work, it is shown that the month-to-month variability of AC over China are dominated by two principal modes: the mono-pole mode and the meridional dipole mode. The first mono-pole mode mainly exhibits the enhanced AC pattern over eastern China. The dipole mode shows two centers over northern and southern China, with positive (negative) values over southern (northern) China. The potential linkages between the two modes and climatic sources are further described. The first mono-pole mode is linked with the 3 months preceding ENSO, and the second dipole mode is connected with the simultaneous NAO.

The possible physical mechanism is also investigated by examining the dynamic and thermal processes involved. For the mono-pole mode, the preceding ENSO can induce anomalous convergence and decrease PBLH over eastern China, which are not favorable for the emission of AC. Meanwhile, it is seen that anomalous negative temperature over eastern China are seen accompanied with the preceding ENSO events, paralleling conditions favorable for enhanced AC. For the meridional dipole mode, anomalous convergence (divergence) and decreased (increased) PBLH are found over northern (southern) China, paralleling the conditions for increased (decreased) AC under the positive phase of NAO. Moreover, the temperature anomalies associated with the NAO over southern and northern China are opposite, agreeing well with the spatial distribution of the dipole mode. That is, both the dynamic and thermal anomalies associated with climate systems are contributed to formation of the leading variabilities of AC over China.

On the other hand, as reported, wet deposition shows important effects in influencing the anomalous distribution of AC (Wu, 2014). However, the role of wet deposition is not discussed in the present work. This is because the influences of ENSO on the seasonal rainfall over China is complex and vary along with the phases of ENSO events. During the decaying summer of a warm ENSO event, above average rainfall is expected to be observed over southern China (e.g., Huang and Wu, 1989; Feng et al., 2016); however, this is not the case for the developing summer (Feng et al., 2016). Moreover, when the intensities of the ENSO events are different, i.e., moderate events vs. strong events, their impacts on the seasonal rainfall over China may vary differently (Xue and Liu, 2008). In addition, it has been indicated that the influence of rainfall on the aerosols exhibits seasonal and regional dependence (Wu, 2014; Feng et al., 2016), and it is found that the role of rainfall is limited in affecting the winter aerosols over southern China (Wu, 2014). However, the month-to-month variability of AC is considered in this study, whereas for a specific season, the potential impacts of wet deposits in determining the distribution of aerosols is complex and uncertainties exist.

In addition, as reported that aerosol has profound effects on climate through aerosol-cloud-radiation interactions, we have further examined the potential impacts of different emissions levels on the distributions of AC. Sensitive experiments are designed by fixing the emissions at the level of year 1986 (low emission) with meteorology field at 1986 and 2006, and at the emissions at the level of year 2006 (high emission) with meteorology field at 1986 and 2006. It is found that even if the emission level is same, the simulated AC are different under different meteorology conditions, suggesting that the role of meteorological conditions in impacting the aerosol concentrations (figure not shown). However, when the anthropogenic emissions have times increased (i.e., emission level at 1986 vs. at 2006), the variation of aerosol concentrations is mainly attributed to the emissions. Due to the limitation of the present study, the relative role of emissions and meteorological conditions on the AC will be discussed in the future work.

Furthermore, the characteristics of the month-to-month variability of aerosols over China is explored, the result highlights the impacts of tropical SST (i.e., ENSO) and the atmospheric system (i.e., NAO or NAM) originating from the Northern Hemisphere on the variability in AC over China. As reported, both ENSO and NAO display considerable influences on the climate anomalies over China (e.g., Huang and Wu, 1989;

Zhang et al., 1996; Gong and Wang, 2003; Li and Wang, 2003), and the result here expands their influences beyond climate. Climate systems, for example, originating from the Southern Hemisphere, display essential influences in affecting seasonal rainfall and temperature anomalies via atmospheric bridges and oceanic bridges (Zheng et al., 2015, 2018). Future work will further examine the potential impacts of the Southern Hemisphere climate systems on the variation in AC over China to comprehensively assess the modulations of climate systems on the AC over China.

Author contribution

JLZ and JF conducted the study design. JLZ performed the simulations. JF and JLZ carried out the data analysis. JPL and HL were involved in the scientific interpretation. JF prepared the manuscript with contributions from all coauthors.

Data availability

The HadISST dataset are downloaded from http://www.metoffice.gov.uk/hadobs/hadisst/data/download.html. The NCEP/NCAR reanalyses is downloaded from http://www.esrl.noaa.gov/psd/data/gridded/. Simulation results and codes to generate figures in this paper have been archived by corresponding authors and are available at https://doi.org/10.5281/zenodo.3247326.

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Figure Captions:

Figure 1. Spatial pattern of the (a) first empirical orthogonal function (EOF1) mode of the monthly surface PM2.5 concentrations over China. (b) As in (a), but for the second mode (EOF2). (c)-(d) As in (a)-(b), but for the column concentrations. The numbers indicate fractional variance in the EOF modes.

Figure 2. (a) The first principal components (PC1) of the monthly PM2.5 concentrations where the red and blue lines are for the surface and column concentrations, respectively. (b) As in (a), but for PC2.

Figure 3. (a) Lead-lag correlation between the Niño3.4 index and PC1. Negative (positive) lags indicate that the Niño3.4 index is leading (lagging) and the dashed lines are the 0.05 significance levels. (b) As in (a), but for the correlation between the NAOI and PC2. The red lines are based on the GEOS-4 meteorological fields, and the blue lines are based on the observations.

Figure 4. Seasonal variations in the standard deviation of the (a) PC1, (b) PC2, (c) Niño3.4 index, and (d) NAOI.

Figure 5. Spatial distribution of the correlation coefficients between the monthly sea surface temperature and PC1 for PC1 lagging for (a) 3 months, (b) 2 months, (c) 1 month, and (d) simultaneous. Color shading indicates significance at the 0.05 level.

Figure 6. Spatial distribution of the correlation coefficients between the monthly sea level pressure and PC2 of the (a) surface and (b) column PM2.5 concentrations. Color shading indicates significance at the 0.05 level.

Figure 7. Spatial distribution of the correlation coefficients between the Niño3.4 index and divergence at 700 hPa for the Niño3.4 index leading for 3 months. Color shading indicates significance at the 0.05 level.

Figure 8. Regressions of the planetary boundary layer height (PBLH) onto the (a) 3-month leading Niño3.4 index and (b) simultaneous NAOI. Color shading indicates significance at the 0.05 level.

Figure 9. Regressions of the convergence at 300 hPa onto the simultaneous NAOI. Color shading indicates significance at the 0.05 level.

Figure 10. Regressions of the skin temperature onto the (a) 3-month leading Niño3.4 index and (b) simultaneous NAOI. Color shading indicates significance at the 0.05 level.