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Aerosol concentrations variability over China: two distinct leading modes

Juan Feng¹, Jianlei Zhu^{2,*}, Jianping Li^{3,4}, and Hong Liao⁵

1. *College of Global Change and Earth System Science, Beijing Normal University, Beijing, China*
2. *Foreign Economic Cooperation Office, Ministry of Ecology and Environment, Beijing, China*
3. *Frontiers Science Center for Deep Ocean Multispheres and Earth System (FDOMES)/Key Laboratory of Physical Oceanography/Institute for Advanced Ocean Studies, Ocean University of China, Qingdao 266100, China*
4. *Laboratory for Ocean Dynamics and Climate, Pilot Qingdao National Laboratory for Marine Science and Technology, Qingdao 266237, China*
5. *Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Jiangsu Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing, 210044, China*

Corresponding author:

Dr. Jianlei Zhu

Foreign Economic Cooperation Office, Ministry of Ecology and Environment, Beijing, China

Email: zhu.jianlei@fecomee.org.cn

Abstract

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

49

50 **1. Introduction**

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

63 Meanwhile, the distribution and accumulation of aerosols are sensitive to
64 meteorological conditions. The variations in the meteorological factors, e.g.,
65 precipitation, wind, temperature, planetary boundary layer height (PBLH), atmospheric
66 stability, and humidity, could impact the AC by modulating the aerosol transport,
67 deposition, and dilution processes (Aw and Kleeman, 2003; Lin and McElroy, 2010;
68 Liao et al., 2015; Yang et al., 2017). The anomalies in meteorological conditions are
69 attributed to the synoptic weather and climate systems. For the synoptic weather scale,
70 Guo et al. (2014) indicated that stagnate weather conditions contribute to the periodic
71 cycle of particulate matter events during boreal winter in Beijing. And the increase in

72 relative humidity (Han et al., 2014) and decrease in the PBLH (Quan et al., 2014; Yang
73 et al., 2015) would lead to an increase in the aerosols, thus contributing to the haze
74 events during winter 2012 in northern China. The variations in the large-scale climatic
75 systems, such as Pacific Decadal Oscillation (PDO), El Niño-South Oscillation (ENSO),
76 East Asian summer and winter monsoon (EASM & EAWM), and North Atlantic
77 Oscillation (NAO) show considerable effects in impacting the regional AC in both the
78 seasonal and the interannual timescales. For example, researchers found that low values
79 of AC are observed in Taiwan accompanied with the onset of the EASM (Chen and
80 Yang, 2008). During the mature phase of the moderate La Niña event 2000/01, an
81 anomalous south-negative-north-positive AC dipole pattern is seen over eastern China
82 (Feng et al., 2017). The interannual variations in the EASM exhibit significant effects
83 in impacting the summertime AC over China, i.e., high-level AC would be observed
84 over eastern China along with a weaker EASM (Zhu et al., 2012; Lou et al., 2016; Mao
85 et al., 2017). A similar situation is observed between the EAWM and AC over eastern
86 China but during boreal winter, showing that a weaker EAWM relates to a high level of
87 AC over China (Jeong et al., 2017). Zhao et al. (2016) have indicated that the decadal
88 regime shift of the PDO showed significant role in impacting the decadal variations of
89 boreal winter aerosols over eastern China. Feng et al. (2019) have reported the
90 important influences of simultaneous ENSO and preceding autumn NAO signals on the
91 winter AC over China by case study.

92 The above discussions highlight the effect of climate background in impacting the
93 AC over China across different seasons, including signals from both the tropical and

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

109 Consequently, one of the crucial motivations of the current work is to investigate
110 the spatial-temporal variability in the monthly AC over China, highlighting the potential
111 effects of climatic variabilities in modulating the spatial and temporal variations in AC,
112 and understanding the possible physical processes involved. The rest of the study is
113 arranged as follows. The model, datasets, and methods are presented in Section 2; The
114 properties of the leading modes of AC variability are described in Section 3; Section 4

115 discusses the contribution of climatic modes on aerosol variabilities; and Section 5
116 provides the conclusions and discussions.

117 **2. Datasets, model, and methodology**

118 **2.1 Model**

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

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

135 The definition of particulate matter smaller than 2.5 μm in diameter (PM_{2.5}) is
136 followed by Liao et al. (2007),

$$137 \quad [PM_{2.5}] = 1.29 \times [NO_3^-] + 1.37 \times [SO_4^{2-}] + [SOA] + [POA] + [BC]$$

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

142 **2.2 Datasets and methodology**

143 The input meteorological variables of the model highly agree with the widely used
144 atmospheric and oceanic datasets, i.e., the National Centers for Environmental
145 Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis
146 (Kalnay et al., 1996), and the UK Meteorological Office Hadley Centre's sea ice and
147 sea surface temperature (SST) datasets (HadISST; Rayner et al., 2003). These two
148 datasets are employed to verify the climatic indices calculated based on the model input
149 meteorological datasets. ENSO was characterized by the Niño 3.4 index, which is
150 defined as the areal averaged SST over 120°W-170°W, 5°N-5°S. The monthly Niño 3.4
151 indices based on the HadISST and model input data are highly related with each other
152 with a correlation coefficient of 0.99, confirming the reliability of the model data. The
153 North Atlantic Oscillation index (NAOI) and Northern Hemisphere Annular Mode
154 index (NAMI) are used to present the sea level pressure (SLP) oscillation between the
155 mid-latitudes and high latitudes in the extratropical Northern Hemisphere. Following

156 Li and Wang (2003), the NAMI is defined as the difference in the normalized global
157 zonal-mean SLP between 35°N and 65°N, in which the 35°N and 65°N refer to the mid-
158 latitude and high latitude, respectively. The definition of the NAOI resembles that of
159 the NAMI but within the North Atlantic sector from 80°W to 30°E. Because the NAOI
160 and NAMI are highly correlated with each other in both spatial distribution and
161 temporal variation (Thompson and Wallace, 1998; Gong et al., 2001), the NAOI is
162 utilized in the current context; similar results are obtained based on the NAMI.

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

169 **3. Distinct leading modes of the variability in aerosol concentrations**

170 **3.1 Two leading modes**

171 Figure 1 presents the spatial distribution of the first (EOF1) and second (EOF2)
172 leading modes based on the monthly surface layer and column AC anomalies. A similar
173 spatial distribution is observed in both the surface and column AC. The EOF1 and EOF2
174 modes explain 31.4% (37.0%) and 16.3% (14.1%) of the total variances for the surface
175 layer (column) AC, respectively. Based on the *North's* rule, the two dominant modes
176 could be significantly separated from each other and from the rest of the eigenvectors

177 based on the analysis of the eigenvalues in the light of sampling error above the 0.05
178 significance level. The rest of the modes are not discussed for their relative less
179 explained variance or could not be well separated. The EOF1 mode displays a mono-
180 sign pattern, with the maximum located in central eastern China (Figs. 1a and c). The
181 EOF2 mode presents a meridional dipole pattern in eastern China, with opposite values
182 to the south (positive values) and north (negative values) of the Yangtze River.

183 The temporal behavior of the two modes, the first and second principal
184 components, i.e., PC1 and PC2, is displayed in Figure 2. Both PC1 and PC2 show strong
185 interannual variations. The PCs based on the surface and column concentrations are
186 closely correlated with each other, with coefficients of 0.80 and 0.79 for PC1 and PC2,
187 respectively. The high consistency between the surface and column concentrations in
188 both the spatial and temporal distributions implies that the factors governing their
189 variations are the same. The maximum value of PC1 occurs in 1998, corresponding to
190 the strongest El Niño event (1997/98) in the 20th century. For PC2, negative values are
191 observed during the winters of 1989 and 2002, and positive values are observed during
192 the winters of 1995 and 1997. However, the winters of 1989 and 2002 correspond to
193 the positive polarities of the NAM or NAO, and the winters of 1995 and 1997 are
194 paralleling to the negative polarities of the NAM or NAO. The potential linkage
195 between the PCs and climatic variabilities is therefore analyzed. Here, the Niño 3.4
196 index is utilized to depict the variation of ENSO, and the NAOI (NAMI) is employed
197 to reflect the variability in the NAO (NAM). Note that the indices based on the model
198 input data are highly correlated with the observation datasets, and the monthly NAOI

199 is closely related with the NAMI, exhibiting a significant correlation coefficient of 0.71
200 during period 1986-2006. Therefore, the NAOI is employed to detect the linkage
201 between the PC2 and climate variability.

202 **3.2 Linkage with the climate variabilities**

203 Figure 3 displays the lead-lag correlation between the PC1 and Niño 3.4 index,
204 and between the PC2 and NAOI to identify the linkage between the climatic
205 variabilities and the two leading AC patterns. PC1 is significantly connected with the
206 Niño 3.4 index, with the maximum occurring when the Niño 3.4 index is 3 months
207 leading, implying a leading influence on PC1. The leading impacts of Niño 3.4 on the
208 variation in PC1 are further seen from the seasonal evolution of the standard deviation
209 in the corresponding indices (Fig. 4). The standard deviation of the monthly Niño 3.4
210 index shows that the maximum occurs during December, while the maximum occurs in
211 March for that of PC1. The leading influences of Niño 3.4 on PC1 are further verified
212 by the spatial distribution of correlations between PC1 and SST, as shown in Figure 5.
213 For the correlation with the PC1 lagged for 3 months, significant positive correlations
214 are observed over the tropical eastern Pacific and Indian Oceans, and negative
215 correlations over the tropical western Pacific. The correlation pattern is like a canonical
216 El Niño pattern. Note that the significant positive correlations over the tropical eastern
217 Pacific gradually decrease as the SST leading time is reduced; however, the correlations
218 over the tropical Indian Ocean become stronger, implying the effects of the Indian
219 Ocean capacitor along with the development of an ENSO event (Xie et al., 2009). The

220 above result ascertains the preceding influence of ENSO on the variation in PC1,
221 indicating a 3-month leading impact of ENSO on the following AC over China.

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

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

240 **4. Physical processes impacting on the leading modes**

241 **5.1 Circulation anomalies associated with ENSO**

242 Figure 7 shows the anomalous circulations associated with ENSO to identify the
243 atmospheric circulation process impacting the EOF1 patterns with the Niño 3.4 index
244 leading for 3 months. It is seen that tropical eastern Pacific and southern China are
245 controlled by significant positive correlations in the correlation with the convergence
246 in the lower troposphere. That is, southern China and tropical eastern Pacific are
247 influenced by anomalous convergence circulation under the influence of a 3-month
248 leading ENSO signal. Meanwhile, tropical western Pacific is impacted by significant
249 negative correlations, indicating that these regions are impacted by anomalous
250 divergence. The anomalous convergence circulation over southern China is not
251 favorable for the transmission of AC. That is the anomalous circulation associated with
252 3-month leading ENSO signal would connect with enhanced AC over eastern China
253 (Feng et al., 2019), which agrees with the spatial distribution of EOF1. Moreover, the
254 impacts of ENSO on the circulation is further seen in impacting the PBLH (Figure 8a).
255 Significant negative anomalies are found over eastern China, indicating that the
256 occurrence of a warm ENSO event would decrease the height of PBLH. The decreased
257 PBLH relates to enhanced AC over eastern China. The above result suggests that the
258 leading ENSO signal exhibits a significant role in affecting the circulation anomalies
259 over China. Under the influence of warm ENSO events, the followed anomalous
260 convergence and decreased PBLH over eastern China are both unfavorable for the
261 emission of AC, contributing to the formation of the EOF1 pattern.

262 **5.2 Circulation anomalies associated with NAO**

263 The anomalous divergence accompanied by the simultaneous NAO is presented in
264 Figure 9. The northern Atlantic Ocean is influenced by an anomalous tri-pole structure,
265 showing convergence-divergence-convergence anomalies from the polar region to the
266 tropical regions. The occurrence of the anomalous circulation structure in the northern
267 Atlantic Ocean is due to the fact that the variation in NAO would induce an anomalous
268 tri-pole SST pattern within the northern Atlantic Ocean (e.g., Wu et al., 2009; Zheng et
269 al., 2016) by which a downstream wave-train is expected to be observed (Ruan et al.,
270 2015; Li and Ruan, 2018). The downstream wave train is seen with significant positive
271 anomalies over southern China in the regression of NAOI to the divergence, while
272 negative anomalies occur over northern China. That is, a positive NAO is accompanied
273 with anomalous divergence (convergence) over southern (northern) China. The
274 anomalous convergence over northern China is unfavorable for the emission of AC,
275 corresponding to enhanced AC. However, the opposite situation is observed over
276 southern China. The anomalous circulation connected with NAO further estimates the
277 negative impacts of NAO on the EOF2 mode.

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

285 over northern and southern China are opposite, consistent with the spatial distribution
286 of the EOF2 mode.

287 **5.3 Role of temperature**

288 Meanwhile, it has been reported that temperature shows an effect in impacting the
289 distribution of aerosols. For example, it is reported that an increase in temperature is
290 associated with a decrease in PM_{2.5} over southern California (Aw and Kleeman, 2003)
291 because enhanced temperature lead to decreases in organics and nitrate (Dawson et al.,
292 2007). Accordingly, the associated impacts of the ENSO and NAO on the temperature
293 over China are detected. Figure 10 displays the anomalous temperature regressed
294 against the 3 months preceding Niño 3.4 index and simultaneous NAOI to detect the
295 temperature anomalies connected with the two climate systems. For a warm event of
296 ENSO, large areas of negative temperature anomalies occupy eastern China, with the
297 maximum lying within the Yellow River and Yangtze River (Fig. 10a). The negative
298 temperature anomalies imply a lower temperature condition, which would induce to
299 enhanced AC.

300 For the NAO, its positive phase corresponds to opposite temperature anomalies
301 over southern and northern China, being positive (negative) over the southern (northern)
302 China (Fig. 10b). Positive temperature anomalies over southern China parallels to a
303 warmer situations and reduced AC in this region. Negative temperature anomalies over
304 northern China set up a background of colder situations, which would increase the AC.
305 The anomalous variation in the temperature agrees with the negative impact of NAO

306 on the AC over eastern China. In addition, the temperature anomalies accompanied with
307 the preceding ENSO are greater than those associated with the simultaneous NAO,
308 highlighting the dominant role of ENSO in impacting the AC over eastern China.

309 **5. Conclusions and Discussions**

310 China has a high loading of aerosols and understanding the variability in AC is
311 important not only for recognizing the interactions between aerosols and climate but
312 also for scientifically understanding the current pollutant status. In the present work, it
313 is shown that the month-to-month variability of AC over China are dominated by two
314 principal modes: the mono-pole mode and the meridional dipole mode. The first mono-
315 pole mode mainly exhibits the enhanced AC pattern over eastern China. The dipole
316 mode shows two centers over northern and southern China, with positive (negative)
317 values over southern (northern) China. The potential linkages between the two modes
318 and climatic sources are further described. The first mono-pole mode is linked with the
319 3 months preceding ENSO, and the second dipole mode is connected with the
320 simultaneous NAO.

321 The possible physical mechanism is also investigated by examining the dynamic
322 and thermal processes involved. For the mono-pole mode, the preceding ENSO can
323 induce anomalous convergence and decrease PBLH over eastern China, which are not
324 favorable for the emission of AC. Meanwhile, it is seen that anomalous negative
325 temperature over eastern China are seen accompanied with the preceding ENSO events,
326 paralleling conditions favorable for enhanced AC. For the meridional dipole mode,

327 anomalous convergence (divergence) and decreased (increased) PBLH are found over
328 northern (southern) China, paralleling the conditions for increased (decreased) AC
329 under the positive phase of NAO. Moreover, the temperature anomalies associated with
330 the NAO over southern and northern China are opposite, agreeing well with the spatial
331 distribution of the dipole mode. That is, both the dynamic and thermal anomalies
332 associated with climate systems are contributed to formation of the leading variabilities
333 of AC over China.

334 On the other hand, as reported, wet deposition shows important effects in
335 influencing the anomalous distribution of AC (Wu, 2014). However, the role of wet
336 deposition is not discussed in the present work. This is because the influences of ENSO
337 on the seasonal rainfall over China is complex and vary along with the phases of ENSO
338 events. During the decaying summer of a warm ENSO event, above average rainfall is
339 expected to be observed over southern China (e.g., Huang and Wu, 1989; Feng et al.,
340 2016); however, this is not the case for the developing summer (Feng et al., 2016).
341 Moreover, when the intensities of the ENSO events are different, i.e., moderate events
342 vs. strong events, their impacts on the seasonal rainfall over China may vary differently
343 (Xue and Liu, 2008). In addition, it has been indicated that the influence of rainfall on
344 the aerosols exhibits seasonal and regional dependence (Wu, 2014; Feng et al., 2016),
345 and it is found that the role of rainfall is limited in affecting the winter aerosols over
346 southern China (Wu, 2014). However, the month-to-month variability of AC is
347 considered in this study, whereas for a specific season, the potential impacts of wet
348 deposits in determining the distribution of aerosols is complex and uncertainties exist.

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

361 Furthermore, the characteristics of the month-to-month variability of aerosols over
362 China is explored, the result highlights the impacts of tropical SST (i.e., ENSO) and the
363 atmospheric system (i.e., NAO or NAM) originating from the Northern Hemisphere on
364 the variability in AC over China. As reported, both ENSO and NAO display
365 considerable influences on the climate anomalies over China (e.g., Huang and Wu, 1989;
366 Zhang et al., 1996; Gong and Wang, 2003; Li and Wang, 2003), and the result here
367 expands their influences beyond climate. Climate systems, for example, originating
368 from the Southern Hemisphere, display essential influences in affecting seasonal
369 rainfall and temperature anomalies via atmospheric bridges and oceanic bridges (Zheng
370 et al., 2015, 2018). Future work will further examine the potential impacts of the

371 Southern Hemisphere climate systems on the variation in AC over China to
372 comprehensively assess the modulations of climate systems on the AC over China.

373

374 ***Author contribution***

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

378 ***Data availability***

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

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559

560 **Figure Captions:**

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

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

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

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

575 **Figure 5.** Spatial distribution of the correlation coefficients between the monthly sea
576 surface temperature and PC1 for PC1 lagging for (a) 3 months, (b) 2 months, (c)
577 1 month, and (d) simultaneous. Color shading indicates significance at the 0.05
578 level.

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

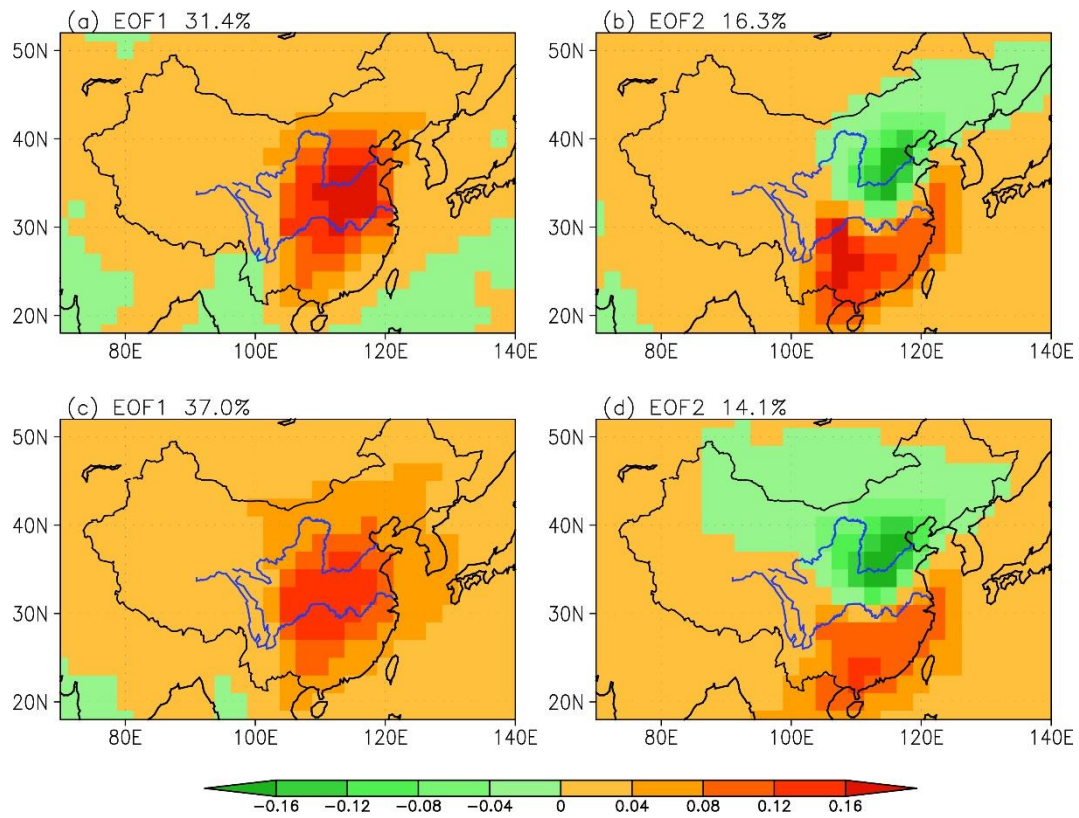
582 **Figure 7.** Spatial distribution of the correlation coefficients between the Niño3.4 index
583 and convergence at 700 hPa for the Niño3.4 index leading for 3 months. Color
584 shading indicates significance at the 0.05 level.

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

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

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

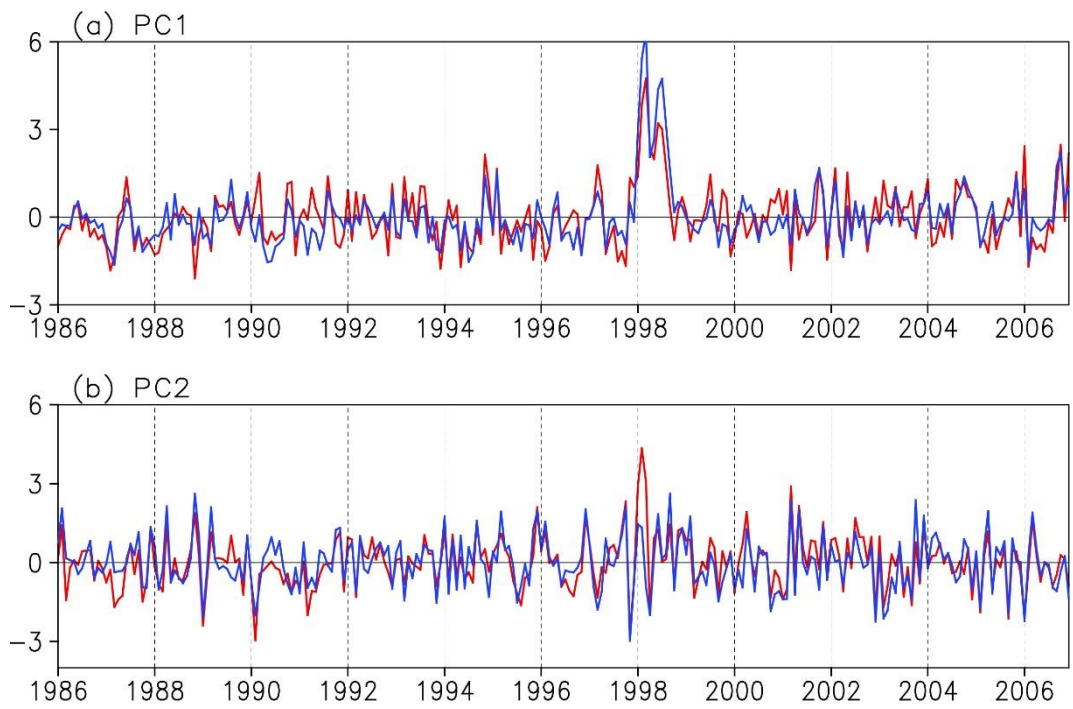
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595 **Figure 1.** Spatial pattern of the (a) first empirical orthogonal function (EOF1) mode of
 596 the monthly surface PM2.5 concentrations over China. (b) As in (a), but for the second
 597 mode (EOF2). (c)-(d) As in (a)-(b), but for the column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

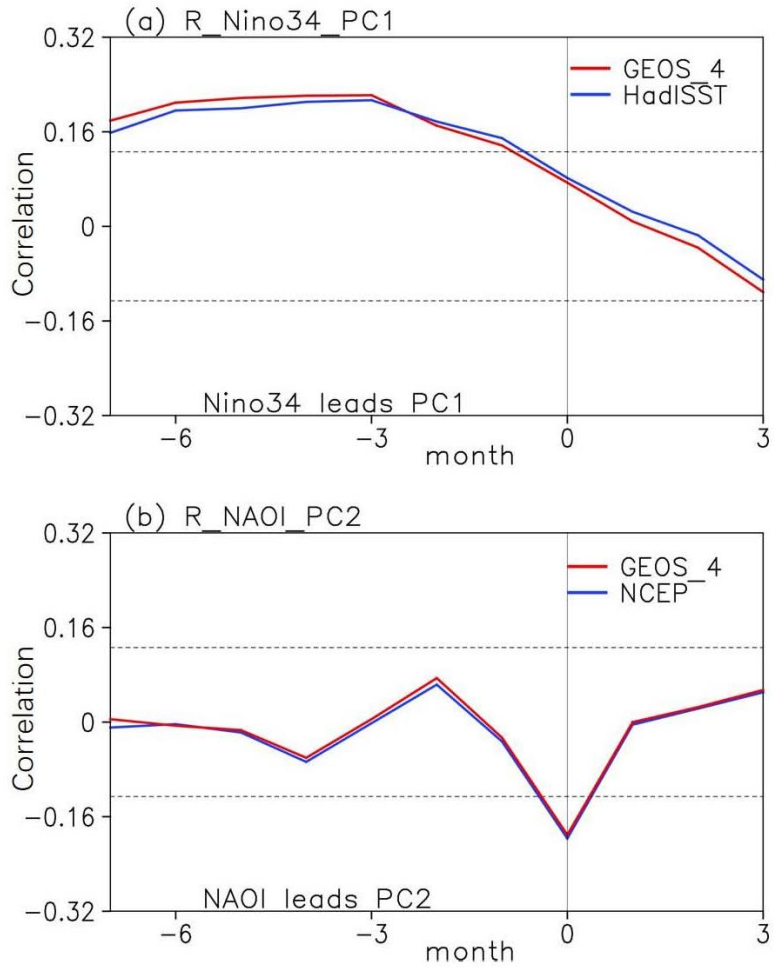
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601 **Figure 2.** (a) The first principal components (PC1) of the monthly PM2.5
 602 concentrations where the red and blue lines are for the surface and column
 603 concentrations, respectively. (b) As in (a), but for PC2.

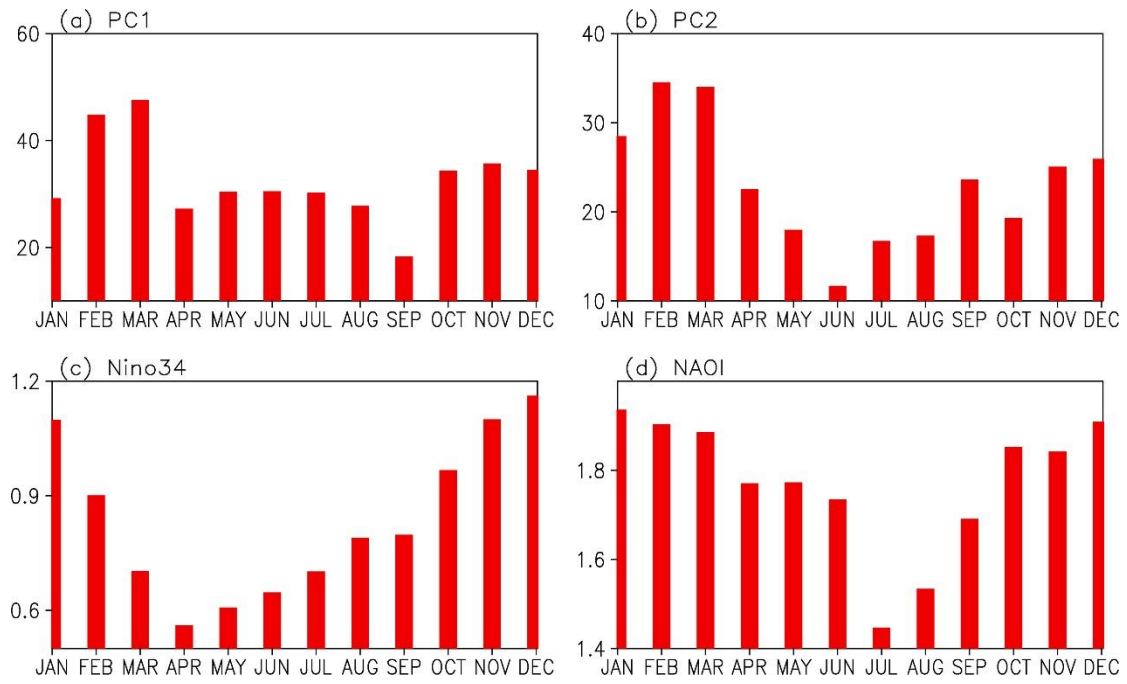
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606 **Figure 3.** (a) Lead-lag correlation between the Niño3.4 index and PC1. Negative
 607 (positive) lags indicate that the Niño3.4 index is leading (lagging) and the dashed lines
 608 are the 0.05 significance levels. (b) As in (a), but for the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

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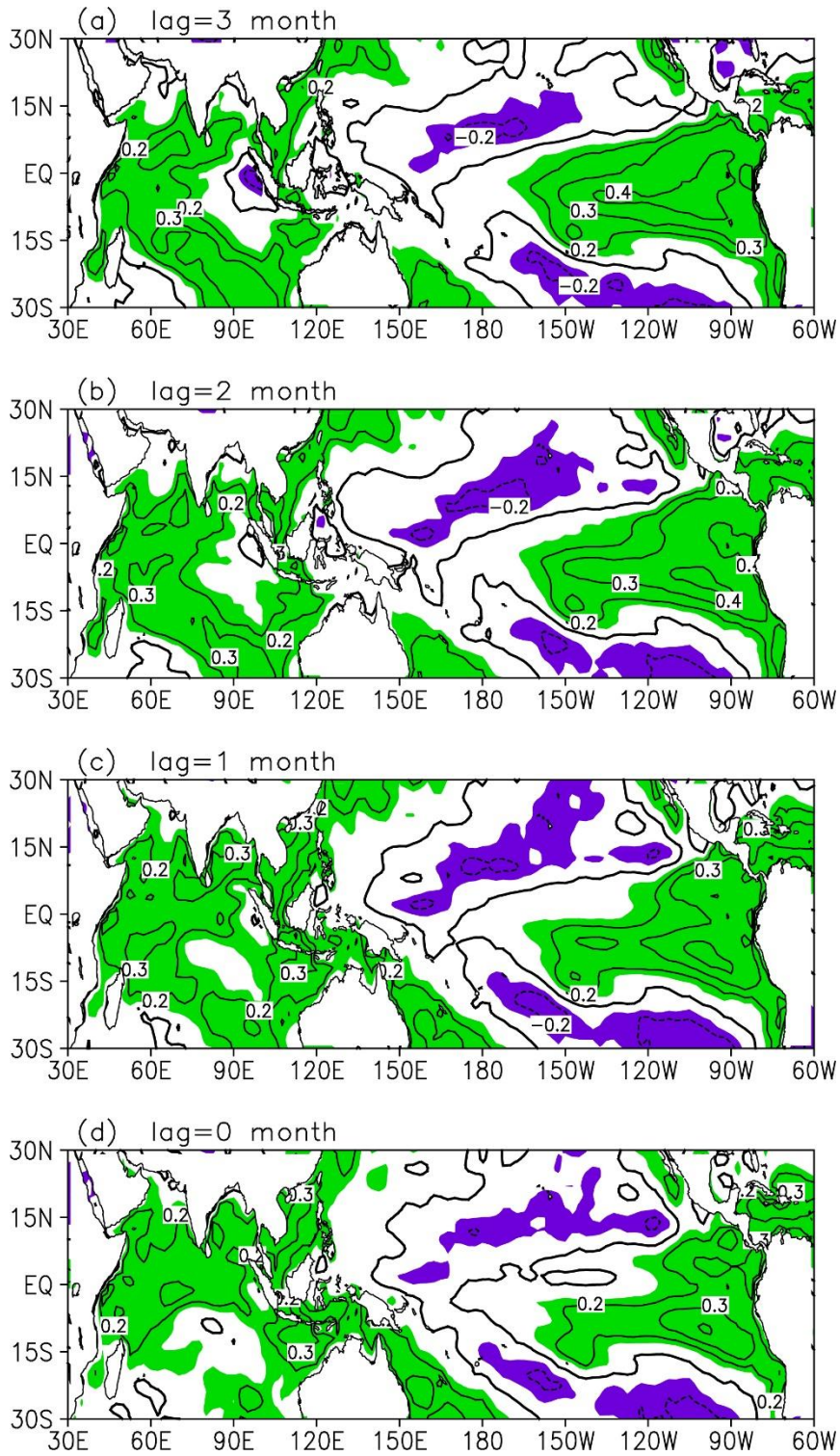


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613 **Figure 4.** Seasonal variations in the standard deviation of the (a) PC1, (b) PC2, (c)

614 Niño3.4 index, and (d) NAOI.

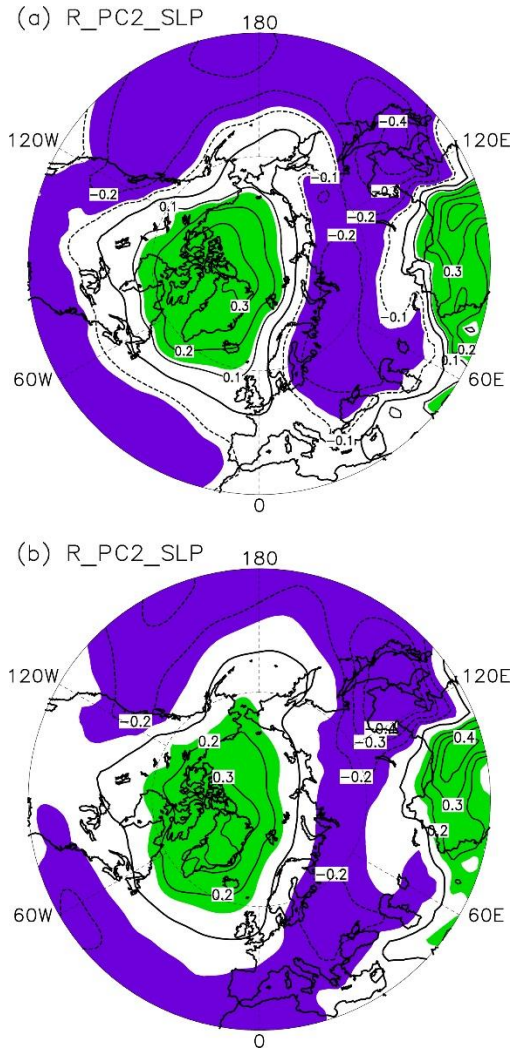
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617 **Figure 5.** Spatial distribution of the correlation coefficients between the monthly sea
 618 surface temperature and PC1 for PC1 lagging for (a) 3 months, (b) 2 months, (c) 1
 619 month, and (d) simultaneous. Color shading indicates significance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

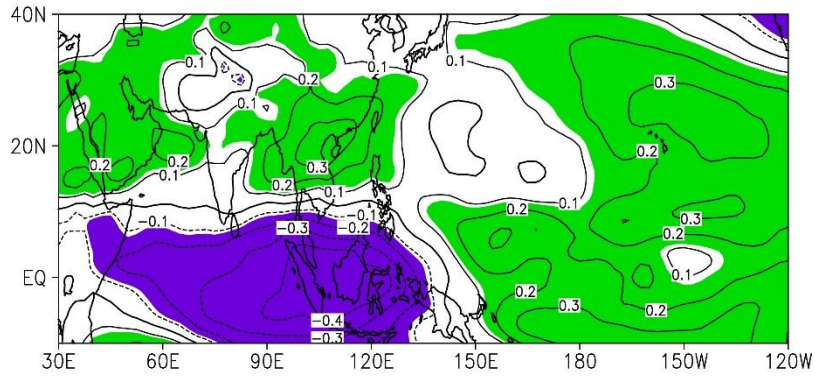
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622 **Figure 6.** Spatial distribution of the correlation coefficients between the monthly sea
 623 level pressure and PC2 of the (a) surface and (b) column PM_{2.5} concentrations. Color
 624 shading indicates significance at the 0.05 level.

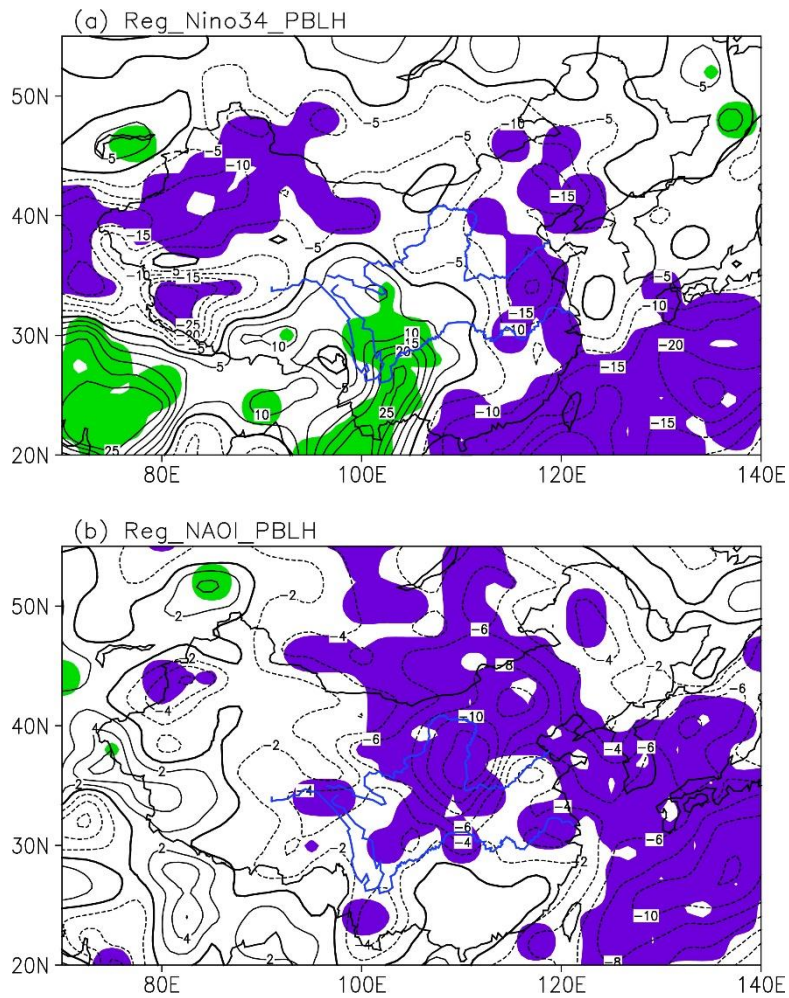
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627 **Figure 7.** Spatial distribution of the correlation coefficients between the Niño3.4 index
 628 and convergence at 700 hPa for the Niño3.4 index leading for 3 months. Color shading
 629 indicates significance at the 0.05 level.

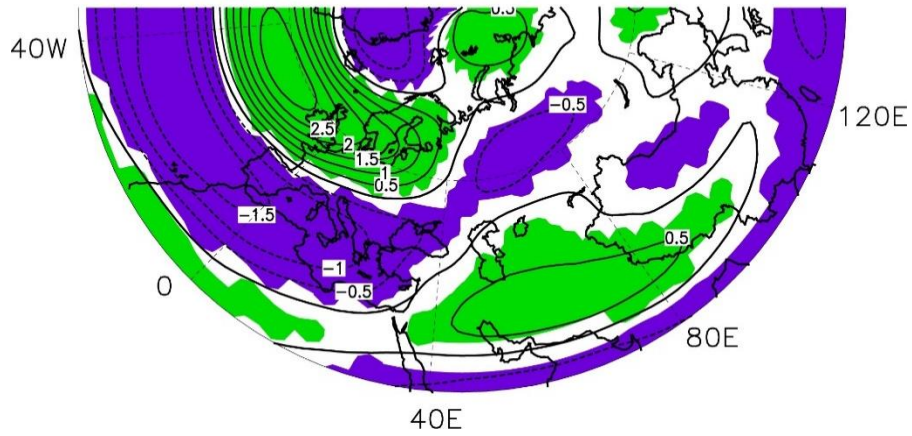
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632 **Figure 8.** Regressions of the planetary boundary layer height (PBLH) onto the (a) 3-
 633 month leading Niño3.4 index and (b) simultaneous NAOI. Color shading indicates
 634 significance at the 0.05 level.

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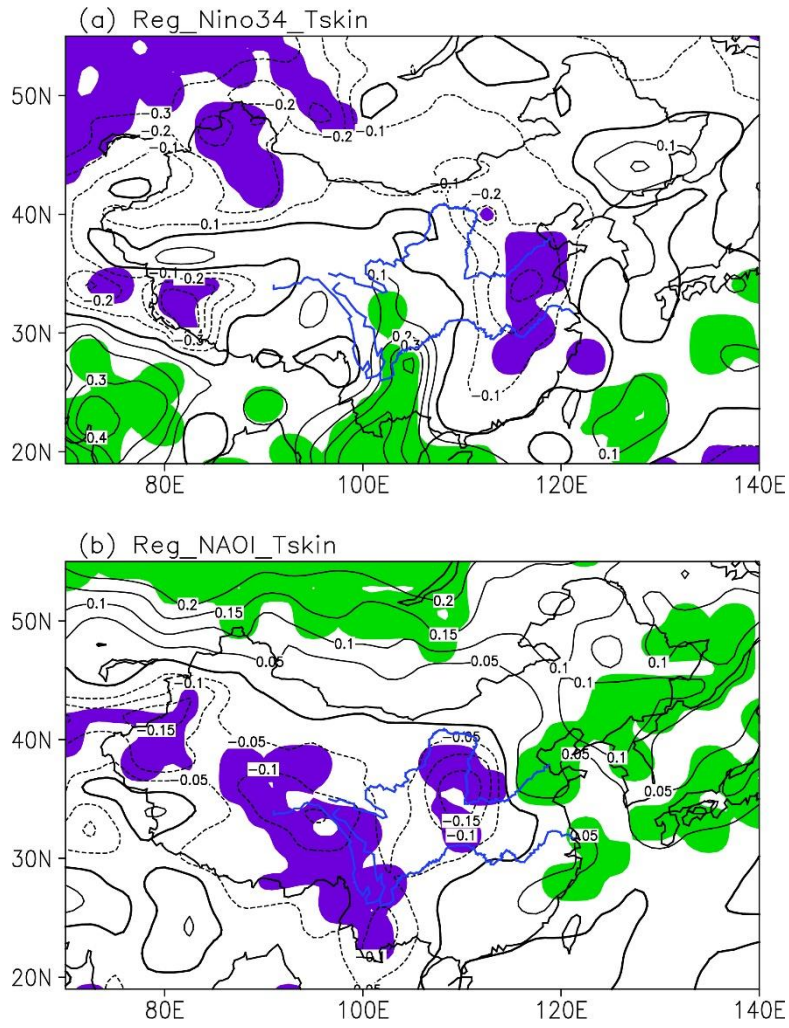


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637 **Figure 9.** Regressions of the convergence at 300 hPa onto the simultaneous NAOI.

638 Color shading indicates significance at the 0.05 level.

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640

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