

RESEARCH ARTICLE

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Key Points:

- The impacts of La Niña Modoki on aerosol concentrations differ between strong (1998/1999) and moderate (2000/2001) events
- The influence of the two La Niña Modoki events on aerosol concentrations may be up to 20% of the climatological mean
- Aerosol concentration anomalies are mainly due to circulation changes associated with the two types of La Niña Modoki rather than wet deposition

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Simulated contrasting influences of two La Niña Modoki events on aerosol concentrations over eastern China

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Abstract Using the Goddard Earth Observing System (GEOS)-Chem model driven by GEOS-4 assimilated meteorological data, the potential influence of two La Niña Modoki events on aerosol concentrations over eastern China is explored. The results indicate that the impact of La Niña Modoki on aerosol concentrations differs between strong and moderate events. During the mature phase of the strong event of 1998/1999, an anomalous dipole pattern is observed over eastern China, i.e., with increased aerosol concentrations in the south and reduced concentrations in the north. The dipole pattern in the moderate event of 2000/2001 is reversed, with reduced aerosol concentrations in the south and increased concentrations in north China. Additionally, decreased aerosol concentrations are seen in spring of the decaying phase for the 1998/1999 event, while in the decaying spring of the 2000/2001 event, the dipole is reversed relative to the mature phase. During the decaying summer, aerosol concentrations are found to increase over eastern China in the 1998/1999 event but not in the 2000/2001 event. These anomalous aerosol concentrations are mainly caused by changes in circulation associated with the two La Niña Modoki events. In contrast, the role of wet deposition is observed to be limited during the lifespan of these two events. It is also found that the potential influence of the two La Niña Modoki events on aerosol concentrations can be up to 20% of the climatological mean. This suggests that La Niña Modoki has an important role in determining the distribution of aerosol concentrations over eastern China.

1. Introduction

Aerosols are an important component of the atmosphere and have a significant influence on the energy budget and air quality of the Earth. Aerosols affect the radiation equilibrium by absorbing or scattering solar radiation [Thompson, 1995] and are able to change the physical and microphysical features of clouds, including their optical properties and precipitation rates [e.g., Hansen et al., 1997; Zhang et al., 2007]. Aerosols thus play an important role in climate. Additionally, aerosols can have adverse impacts on human health [e.g., Dockery et al., 1993; Pope et al., 1995; Tie et al., 2009], atmospheric visibility [Watson, 2002], and environmental pollution [Intergovernmental Panel on Climate Change, 2007]. Therefore, a better understanding of the spatiotemporal variability and distribution of aerosols would benefit both scientific and social endeavors.

Aerosols are both directly emitted into the atmosphere and created in situ through the chemical production of secondary aerosols. These processes are sensitive to atmospheric variability. Changes in the climate could affect aerosol concentrations in many ways, such as through wind transportation, the perturbation of ventilation rates, wet deposition, variation in natural emissions, and dry deposition. Therefore, understanding the effects of climate variation on regional air quality is of key importance to future air quality planning.

The influence of atmospheric circulation on aerosol concentrations has been widely discussed; wind [Zhang et al., 2010; Zhu et al., 2012; Feng et al., 2016a], temperature [Aw and Kleeman, 2003; Dawson et al., 2007], humidity [Tai et al., 2010; Ding and Liu, 2014], and atmospheric boundary layer thickness [Kleeman, 2008; Zhang et al., 2014; Yang et al., 2016] have considerable influence on the spatiotemporal variability and distribution of aerosol concentrations. Previous studies have reported that the seasonal variations of

aerosol concentrations in Hong Kong [Tan *et al.*, 1998] and Taiwan [Chen and Yang, 2008] are related to the timing of the monsoon onset. Variations in the Asian summer monsoon affect the vertical transport of aerosols via deep convection and upper tropospheric anticyclones [e.g., Gettelman *et al.*, 2004; Randel *et al.*, 2010; Zhang *et al.*, 2010]. Additionally, the interannual variations of aerosol concentrations over eastern China are connected to the variability of the East Asian summer monsoon (EASM) [Zhang *et al.*, 2010; Zhu *et al.*, 2012]. Subsequently, Zhu *et al.* [2012] reported that the decadal-scale weakening of the EASM resulted in increased aerosol concentrations over northern China. Thus, the role of the atmospheric circulation in determining aerosol concentrations cannot be ignored.

Previous studies have shown that both the warm (El Niño) and cold (La Niña) events of El Niño–Southern Oscillation (ENSO) have significant impacts on circulation and rainfall anomalies over eastern China, via their modulation of the shift of the western Pacific subtropical high [e.g., Huang and Wu, 1989; Zhang *et al.*, 1999; Feng and Li, 2011; Yuan and Yang, 2012; Feng *et al.*, 2016b]. For example, positive rainfall anomalies occur over southern China during the decaying spring of El Niño events [e.g., Feng and Li, 2011], but positive summer rainfall anomalies over the Yangtze and Huai-He River valleys and southern China and negative rainfall anomalies over northern China are observed during the developing autumn of El Niño events [Feng *et al.*, 2016b]. Recently, a new variant of the El Niño phenomenon has been identified, associated with sea surface temperature anomalies (SSTAs) in the central Pacific. This phenomenon is associated with a tripolar pattern of sea level pressure anomalies during its evolution and two anomalous Walker circulation cells in the troposphere. A typical ENSO has only a single cell. This phenomenon is referred to as the date line El Niño [Larkin and Harrison, 2005], El Niño Modoki [Ashok *et al.*, 2007], central Pacific El Niño [Yu and Kao, 2007; Kao and Yu, 2009], or warm pool El Niño [Kug *et al.*, 2009]. Moreover, it is found that this new type of El Niño has important teleconnections and regional climatic effects, which are different from those associated with the canonical El Niño [e.g., Weng *et al.*, 2007, 2009; Feng and Li, 2013]. Recently, increased aerosol concentrations were reported during the mature phase of the 1994/1995 (boreal winter) El Niño Modoki event, whereas decreased anomalies were found during its decaying phase from boreal spring to autumn [Feng *et al.*, 2016a].

The El Niño events of the two types of ENSO have been widely discussed, but few studies have investigated La Niña events. This is because it is still a matter of debate whether the La Niña events can be divided into two different types just like El Niño events. Some studies showed no obvious change of the zonal location of the maximum SSTA center between individual La Niña events [e.g., Kug *et al.*, 2009; Ren and Jin, 2011], while some other studies have argued for the existence of two types of La Niña event [e.g., Cai and Cowan, 2009; Zhang *et al.*, 2015]. However, there are two different types of teleconnection produced by La Niña, with associated regional climate effects [Feng and Li, 2011; Karori *et al.*, 2013]. These two impacts of La Niña provide indirect evidence for the existence of the two types of La Niña. In addition, Kug and Ham [2011] reported that there is an asymmetry between El Niño and La Niña events for both types of ENSO, with La Niña events being less distinctive than El Niño events. Wang *et al.* [2012] reported that the tropical cyclone activity in the western North Pacific might be affected by the two types of ENSO and found that La Niña events play a particular role in influencing the lifetime of tropical cyclones. Zhang *et al.* [2015] found that the SSTA evolution of the two types of La Niña event is different and the two types of La Niña are associated with different teleconnections; i.e., a negative North Atlantic Oscillation (NAO) and weakening Atlantic jet are linked with the canonical La Niña. In contrast, a positive NAO-like climate anomaly is observed with a strengthening Atlantic jet associated with La Niña Modoki events. Therefore, it is worthwhile to further examine the possible influences of the cold events of the two types of ENSO on the climate.

Meanwhile, Xue and Liu [2007] reported that enhanced rainfall is observed over the Yangtze River valley and decreased rainfall over north China during the decaying summer of strong El Niño events. This is consistent with the flooding that occurred in the summers of 1983, 1998, and 2016, which are the decaying summers of three strong El Niño events. However, there is increased rainfall over north China during the decaying summer of moderate El Niño events. These results indicate that the impact of ENSO on climate over China varies with ENSO intensity and that the influence of strong El Niño events is different from that of moderate events. China currently experiences a relatively high aerosol loading, particularly in eastern China [Gao, 2008; Che *et al.*, 2009; Yang *et al.*, 2015; Feng *et al.*, 2016c]. It is therefore important to examine the potential influence of La Niña events and the potential differences between strong and moderate La Niña events on the aerosol concentrations over eastern China.

In this study, only the potential influence of La Niña Modoki on the aerosol concentrations over eastern China is explored. There are three main reasons for this. (1) There were two moderate canonical La Niña events during the period of 1986–2006 when the simulations of Goddard Earth Observing System chemical transport model (GEOS-Chem) are available, i.e., 1995/1996 and 2005/2006. However, the values of the Niño3 index based on the GEOS-4 fields and the Met Office Hadley Centre's sea ice and SST data sets (HadISST) [Rayner *et al.*, 2003] during the mature phase of the two events show certain differences, i.e., -0.75°C versus -0.63°C for 1995/1996 and -0.69°C versus -0.85°C for 2005/2006. Considering that a La Niña event itself involves strong air-sea interactions and is sensitive to the intensity of the SSTA, the uncertainty in the strength of these La Niña events makes it difficult to draw conclusions from their study. (2) The decaying summer of the 1995/1996 canonical La Niña event was accompanied by a weak EASM (summer 1996), whereas the summer of 2006 was accompanied by a normal EASM. The strength of the EASM has been found to have an important influence on the aerosol concentrations over China [Zhang *et al.*, 2010; Zhu *et al.*, 2012]. (3) The winter of 1995/1996 coincided with a strong East Asian winter monsoon (EAWM), whereas there was a normal EAWM in the winter of 2005/2006 [Wu and Wang, 2002]. However, the variation of the EAWM is connected with the variability of ENSO [Wang *et al.*, 2009], and the EAWM also has an important influence on aerosol concentrations over China [Zhang *et al.*, 2014]. Thus, it would be difficult to separate the influences of EAWM and La Niña during the winter of 1995/1996. Therefore, it is likely that the impacts of EASM and La Niña would also combine. The above three considerations suggest that these two La Niña events should be discounted and make it difficult to explore the possible influences of canonical La Niña on the aerosol concentrations over China with the currently available data sets.

Meanwhile, we reported previously that the El Niño Modoki plays an essential role in influencing aerosol concentrations during its lifespan and the impacts vary between developing and decaying phases [Feng *et al.*, 2016a]. In this study we explore the possible impacts of its counterpart, the La Niña Modoki, on the aerosol concentrations, to gain a better understanding of the influence of ENSO Modoki on aerosol concentrations over China. Section 2 describes the model and data sets used in this study. We investigate the possible impacts of La Niña Modoki on seasonal aerosol concentrations over eastern China in section 3. We characterize the possible physical processes involved and determine the relative roles of circulation and rainfall anomalies associated with La Niña Modoki on aerosol concentrations in section 4. Discussions and conclusions are provided in section 5.

2. Model and Data Sets

A three-dimensional tropospheric chemistry model, the GEOS-Chem (version 8.02.01; http://acmg.seas.harvard.edu/geos/geos_overview.html) [see also Bey *et al.*, 2001], is used to simulate the aerosol distribution. This model is driven by assimilated meteorological observations from the NASA Global Modeling and Assimilation Office GEOS model. All the simulations are driven by GEOS-4 meteorological fields on a horizontal grid with a resolution of 2° latitude by 2.5° longitude. The aerosol deposition follows Wesely [1989] for dry and Liu *et al.* [2001] for wet deposition. Global emissions of ozone precursors, aerosol precursors, and aerosols in the GEOS-Chem model are the same as those described in Zhu *et al.* [2012] and Feng *et al.* [2016a]. The detailed coupled treatment of tropospheric ozone- NO_x -hydrocarbon chemistry, aerosols, and their precursors is the same as in previous studies [e.g., Bey *et al.*, 2001; Park *et al.*, 2003, 2004; Liao *et al.*, 2007]. To discuss the possible influences that La Niña Modoki events may have on aerosol levels, concentrations were expressed as $\text{PM}_{2.5}$, according to Liao *et al.* [2007], and defined as

$$[\text{PM}_{2.5}] = 1.37 \times [\text{SO}_4^{2-}] + 1.29 \times [\text{NO}_3^-] + [\text{BC}] + [\text{POA}] + [\text{SOA}],$$

where the factors 1.37 and 1.29 are used for converting measured sulfate (SO_4^{2-}) and nitrate (NO_3^-) ions to concentrations of ammonium sulfate and ammonium nitrate, respectively, following the factors used for the Interagency Monitoring of Protected Visual Environments [Malm *et al.*, 1994]. Natural aerosols (i.e., dust and sea salt aerosols) are not considered in the calculation of $\text{PM}_{2.5}$ but are simulated in the model. The simulation period was 1986–2006, corresponding to the input data sets available. To isolate the possible impacts of circulation and external emissions on aerosol concentrations, the anthropogenic emissions and biomass burning emissions were fixed in the simulations. Therefore, the variations of aerosol concentrations are due to the impacts of meteorological conditions related to climatic events.

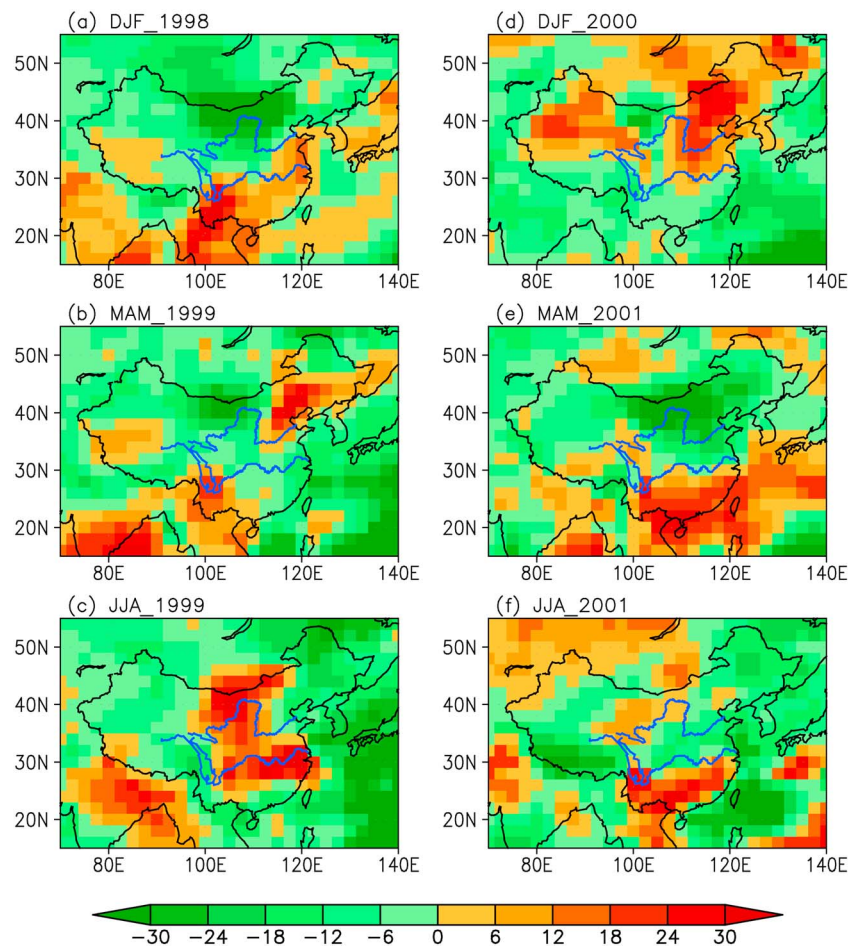


Figure 1. Spatial distribution of the surface-layer PM_{2.5} RPAD during the seasonal cycle for the La Niña Modoki events of (left) 1998/1999 and (right) 2000/2001.

The ENSO Modoki index (EMI) is defined following *Ashok et al. [2007]*, as

$$EMI = [SSTA]_c - 0.5 \times [SSTA]_E - 0.5 \times [SSTA]_W,$$

where the square brackets with a subscript represent the area mean SSTA over the central Pacific region (C: 165°E–140°W, 10°S–10°N), eastern Pacific region (E: 110°–70°W, 15°S–5°N), and western Pacific region (W: 125°–145°E, 10°S–20°N), respectively. According to *Zhang et al. [2015]* there are three well-defined La Niña Modoki events during the period of 1986–2006, i.e., 1988/1989, 1998/1999, and 2000/2001. We have used the skin temperature to calculate the EMI. The EMI here is highly correlated with the EMI based on HadISST (with a correlation coefficient of 0.96 over the period of 1986–2006) [*Feng et al., 2016a*]. Note that the mature phase values of EMI (mean of December–January–February) based on HadISST are very different from those using GEOS-4 for the event of 1988/1989 (–1.07°C versus –1.25°C) and the La Niña Modoki event includes strong air-sea interactions. In addition, the EMI during 1988/1989 lies between the EMI for the 1998/1999 event (with mature phase EMI value of –1.27°C) and that for 2000/2001 (–0.69°C). In this study, we aim to explore the different influences of strong and moderate La Niña Modoki events. For this purpose, the events of 1998/1999 and 2000/2001 may be more representative than the 1988/1989 event. Therefore, the 1988/1989 event has been discounted in this study. An event with EMI value below –1°C is defined as a strong La Niña Modoki event and between –1°C and –0.5°C as a moderate La Niña Modoki event. Accordingly, the event of 1998/1999 corresponds to a strong La Niña Modoki event, whereas the event of 2000/2001 is classed as a moderate La Niña Modoki event. The differences between these strong and moderate La Niña Modoki events can be compared. In addition, both the winters of 1998/1999 and 2000/2001 coincide with normal EAWM events [*Wu and Wang, 2002*] and the summers of 1999 and 2001

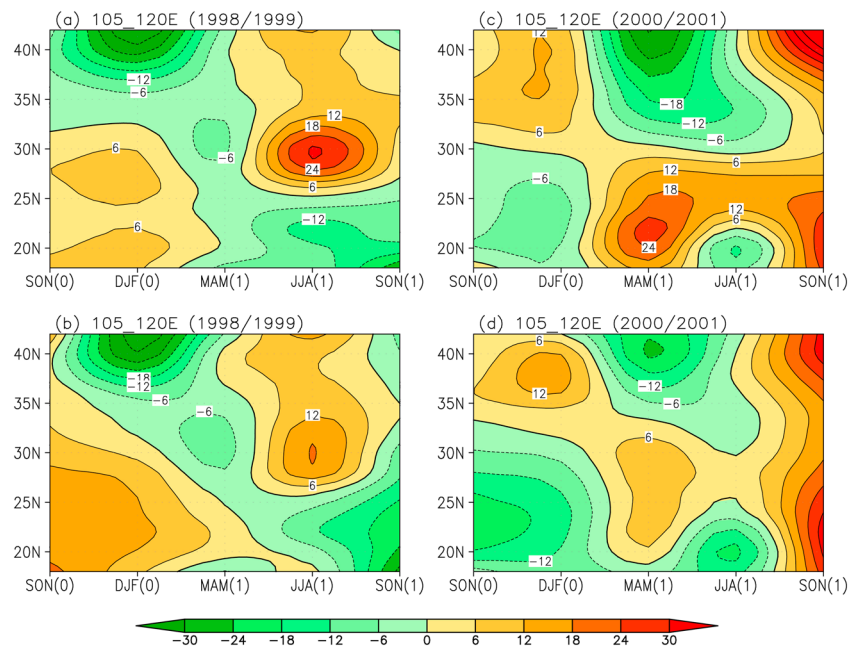


Figure 2. (a) As in Figure 1, but for the latitude-time cross section averaged over the longitude range of 105°–120°E during the La Niña Modoki event of 1998/1999. September–October–November (SON(0)) and December–January–February (DJF(0)) refer to the SON and DJF in 1998, and March–April–May (MAM(1)), June–July–August (JJA(1)), and SON(1) refer to 1999. (c) As in Figure 1a, but for the La Niña Modoki event of 2000/2001. (b and d) The distribution of column-integrated abundance of PM_{2.5} RPAD.

are associated with normal EASM events [Zhu *et al.*, 2012]. That is, the magnitudes of EAWM and EASM in the two events are equivalent, so the two events may reflect mainly the influence of La Niña Modoki rather than a mixed role from the monsoons.

To quantify the influences of the La Niña Modoki on the aerosol concentrations, the Relative Percentage of Anomalies Departure (RPAD) from the climatological mean during the La Niña Modoki events is defined as

$$RPAD(i, j, k) = 100\% \times \left(P(i, j, k) - \frac{1}{n} \sum_{m=1}^n P(i, j, m) \right) / \frac{1}{n} \sum_{m=1}^n P(i, j, m),$$

where $P(i, j, k)$ is the simulated seasonal mean aerosol concentrations at grid point (i, j) of year k and n is the number of years examined (i.e., in this study $n = 21$ to represent 1986–2006).

The input meteorological fields of the GEOS-Chem model have previously been shown to be highly consistent with widely used reanalyses [Zhu *et al.*, 2012; Feng *et al.*, 2016a], such as the National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis [Kalnay *et al.*, 1996]. Moreover, the GEOS-Chem model has been widely used in the atmospheric sciences and beyond and proven to be a useful tool for understanding the variability of aerosol concentrations. The model performance within East Asia has been evaluated in many recent studies [e.g., Fu and Liao, 2014; Mu and Liao, 2014; Lou *et al.*, 2014; Feng *et al.*, 2016c] that have shown that the seasonal and interannual variations of aerosol concentrations and ozone over eastern China are well captured in GEOS-Chem [e.g., Wang *et al.*, 2011; Yang *et al.*, 2014; Feng *et al.*, 2016a].

3. Aerosol Anomalies Associated With the Two La Niña Modoki Events

The spatial distributions of RPAD for PM_{2.5} during the two La Niña Modoki events of 1998/1999 and 2000/2001 are displayed in Figure 1. For the strong La Niña Modoki event of 1998/1999, the aerosol concentration anomalies show a dipole structure over eastern China during its mature winter phase, i.e., positive in the south and negative in the north, divided by the Yangtze River around 30°N (Figure 1a). However,

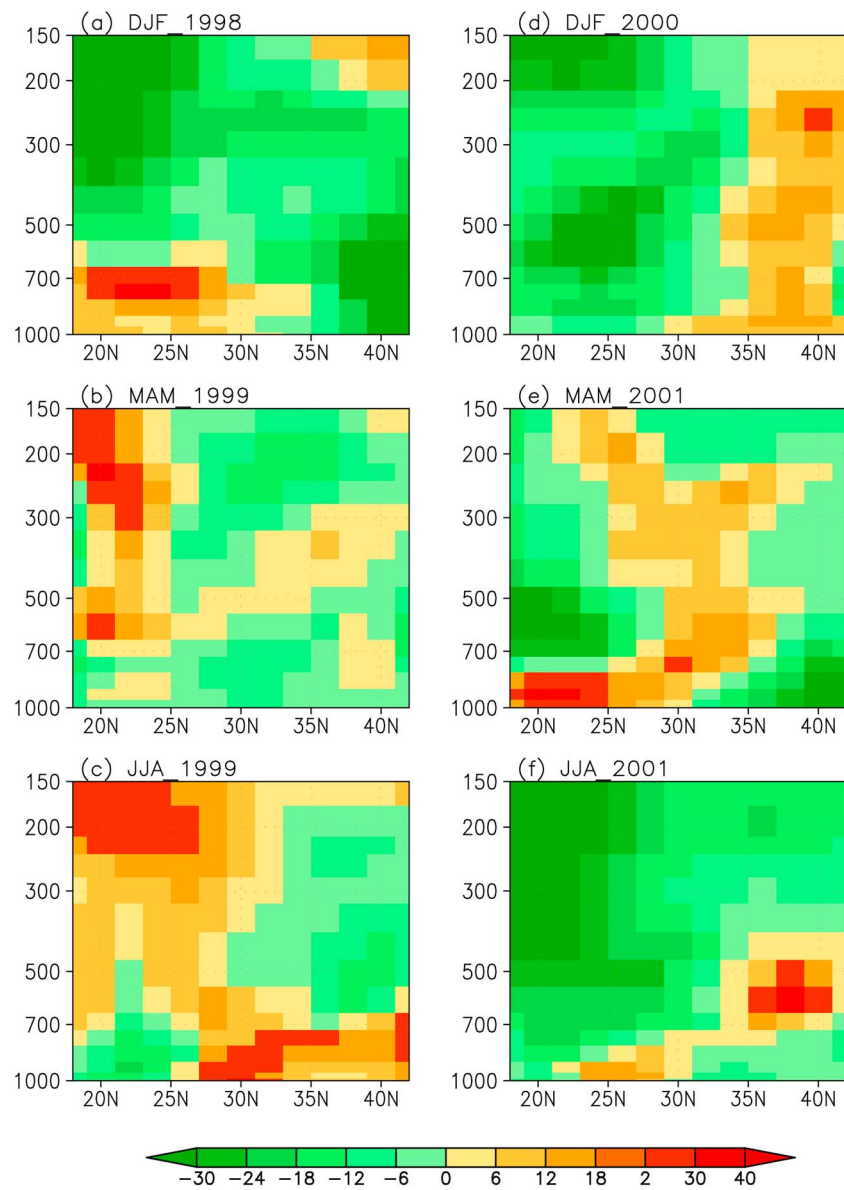


Figure 3. As in Figure 1, but for the pressure-latitude distribution of zonally averaged surface $PM_{2.5}$ RPAD over 105° – 120° E during the La Niña Modoki events of (left) 1998/1999 and (right) 2000/2001.

the positive anomalies in the south are largely reduced in the following spring and the negative anomalies in the north are also weakened (Figure 1b). From the decaying phase spring to summer, the aerosol concentrations over eastern China continuously increase, with positive anomalies in the regions north of 25° N and negative anomalies in the southern coastal regions (Figure 1c). Similar characteristics are also seen in the column concentrations (figures not shown). The strong La Niña Modoki event of 1998/1999 results in a dipole aerosol distribution in eastern China, which later changes to mainly positive anomalies. The dipole pattern during the mature phase in the winter during the La Niña Modoki event of 1998/1999 would worsen air quality conditions in southeast China but improve air quality conditions in northeast China. In contrast, the positive anomalies during the decaying phase summer would worsen air quality conditions in the La Niña Modoki event of 1998/1999.

As for the moderate La Niña Modoki event of 2000/2001, a contrasting dipole distribution is observed in the mature phase over eastern China (Figure 1d), with negative anomalies in the south and positive anomalies in the north. Strong and moderate La Niña Modoki events can therefore be distinguished from each

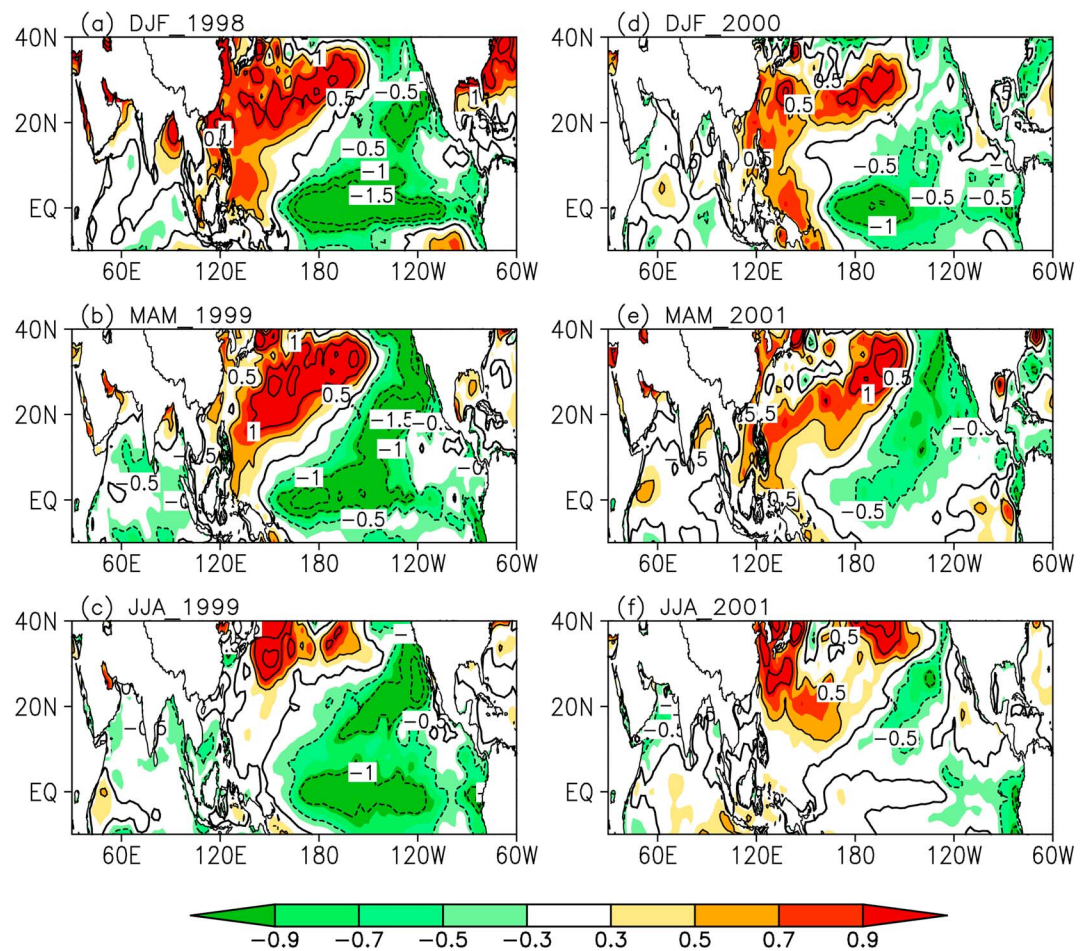


Figure 4. Horizontal distribution of SSTA ($^{\circ}\text{C}$) based on the assimilated meteorological data during the La Niña Modoki events of (left) 1998/1999 and (right) 2000/2001.

other. Additionally, there is a significant spatial correlation coefficient of -0.30 between the strong and moderate events of aerosol concentrations within the region of $105^{\circ}\text{--}120^{\circ}\text{E}$, $20^{\circ}\text{--}40^{\circ}\text{N}$. However, the dipole structure is reversed in the following spring 2000/2001, with maximum values reaching approximately 30% (Figure 1e). The spatial correlation coefficient between the mature winter and decay spring distributions is -0.81 , indicating opposite effects in the mature winter and decaying spring phases of the La Niña event of 2000/2001. The dipole pattern of the decaying phase spring remains until the summer, albeit with reduced absolute values (Figure 1f). The dipole structure during the mature phase of the La Niña Modoki event of 2000/2001 would worsen the air quality conditions in northeast China but would improve the air quality conditions in southeast China.

The above features of the aerosol variations during the two La Niña Modoki events are further established in Figures 2 and 3. Moreover, similar features are seen in the aerosol column concentrations (Figures 2c and 2d). This suggests that the aerosol concentrations are mainly located near the surface.

Both La Niña Modoki events influence aerosol concentrations over eastern China, affecting aerosol concentrations, spatial distributions, seasonal evolution, and occurrence of anomalous peak values. Although the peak anomalies occurred in the decaying summer of the La Niña Modoki event of 1998/1999 (Figures 2a and 3c), for the La Niña Modoki event of 2000/2001, they occurred in the decaying spring (Figures 2c and 3e). The results thus indicate that the La Niña Modoki has a considerable impact on aerosol concentrations over eastern China and highlights the importance of discriminating the intensity of La Niña Modoki because of the potential opposite influences of strong and moderate events on the aerosol concentrations.

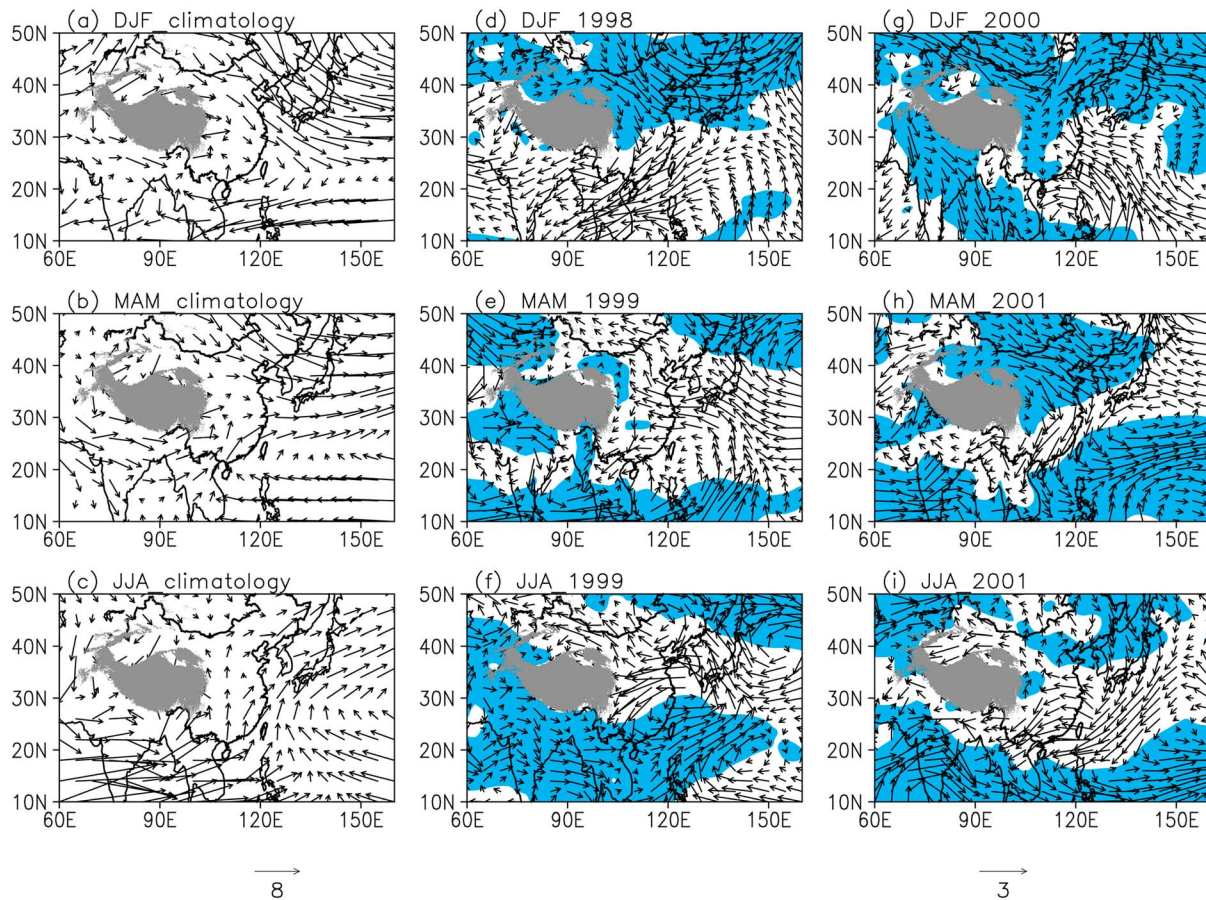


Figure 5. Horizontal distribution of (left) climatological mean wind at 850 hPa and the wind anomalies (vectors; m s^{-1}) and divergence (shading for positive values; s^{-1}) at 850 hPa during the La Niña events of (middle) 1998/1999 and (right) 2000/2001.

4. Mechanisms of the Impact of La Niña Modoki Events on Aerosol Concentrations

The results above indicate that La Niña Modoki events have considerable impact on aerosol concentrations over eastern China. The possible physical processes involved will be illustrated in this section. The underlying temperature (Figure 4) and circulation anomalies (Figure 5) associated with the two La Niña Modoki events are examined. Note that the spatial distribution of wind anomalies at 850 hPa during the two La Niña Modoki events based on the GEOS-Chem input meteorological fields and those from NCEP/NCAR (figures not shown) show similar spatial structures, supporting the reliability of the analysis.

During the mature phases of the two events, we see that the SSTAs over the central tropical Pacific in the La Niña Modoki event of 1998/1999 are much larger and extend farther westward than those in the La Niña Modoki event of 2000/2001 (Figures 4a and 4d). The maximum difference in the SSTA between the two La Niña Modoki events is above 0.7°C in the central tropical Pacific during boreal winter (figure not shown). Divergence occurs in response to the anomalous cooling conditions, with anomalous southerlies/southeasterlies in the western Pacific to the north of the equator (Figures 5d and 5g). Due to the location and intensity of the SSTA in the two events, the anomalous divergent circulation for the 1998/1999 event shifts northwestward compared with its location during the 2000/2001 event (Figures 5d and 5g). In the western Pacific, the anomalous winds would oppose the climatological wind (Figure 5a), which is associated with positive SSTA (Figures 4a and 4d). Note that the extent of the SSTA in the western Pacific is consistent with the SSTA in the central Pacific; i.e., the anomalies of the 1998/1999 event are much stronger and located farther north than those of the 2000/2001 event. Due to the anomalous positive SSTA in the western Pacific, cyclonic rotational flow occurs over the southern China coastal regions. However, the locations of the cyclonic flows are different in the two events due to different Rossby wave response patterns

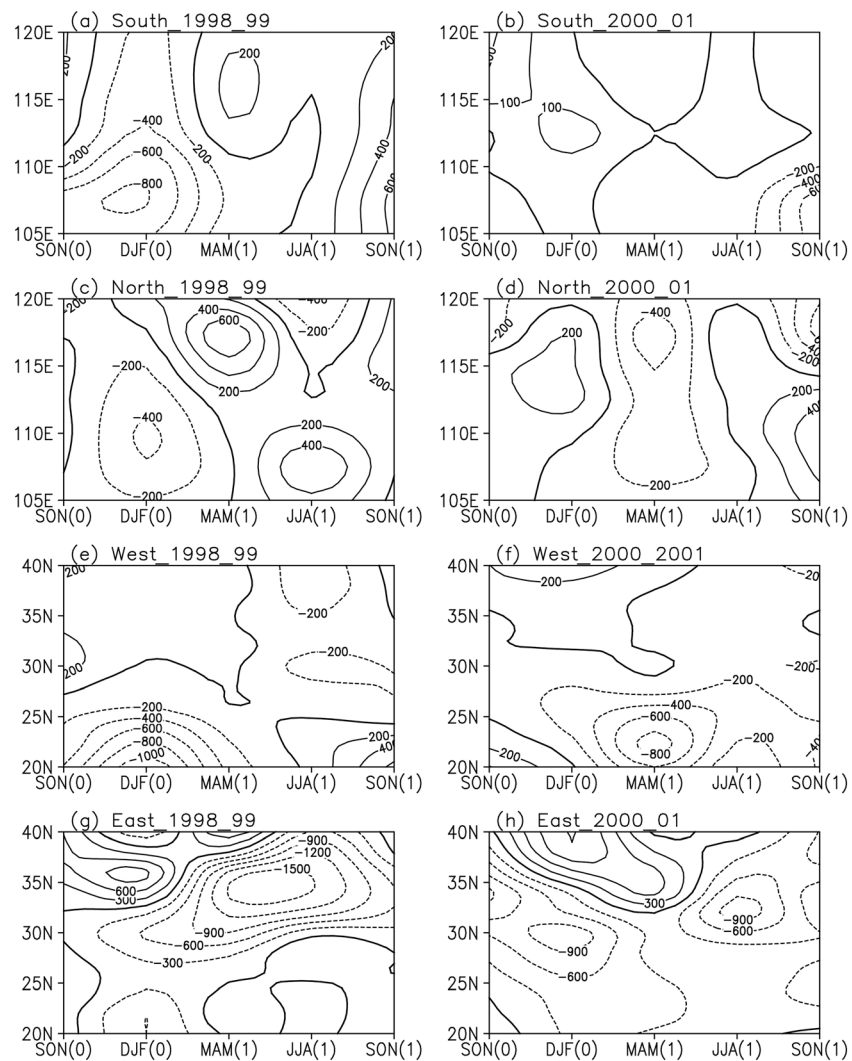


Figure 6. Anomalies of vertically integrated aerosol concentrations fluxes at the (a and b) south, (c and d) north, (e and f) west, and (g and h) east boundaries of eastern China from 1000 to 100 hPa during the La Niña events of (left) 1998/1999 and (right) 2000/2001.

induced by the cooling in the central Pacific. For the 1998/1999 event, the anomalous cyclonic circulation is located over the southwest of south China and south China is controlled by anomalous convergence (Figure 5d). This situation is not favorable for the dispersion of aerosols, resulting in increased aerosol concentrations in the winter of the 1998/1999 event (Figure 1a). In contrast, in the 2000/2001 event, the anomalous cyclonic flow is over the southeast of south China and south China is influenced by anomalous southeasterlies (Figure 5g). This would bring clean air into south China, resulting in reduced aerosol concentrations over south China during the mature phase of the 2000/2001 event (Figure 1d). Meanwhile, the occurrence of positive SSTA in the northern Pacific during the La Niña Modoki event would intensify the convergence there. Similarly, due to the different locations of the warm SSTA in the northern Pacific associated with the two events (Figure 4a versus Figure 4d), the anomalous cyclonic circulation in the event of 1998/1999 is farther north than in the event of 2000/2001 (Figure 5d versus Figure 5g). Consequently, there are anomalous northwesterlies in north China during the winter of the 1998/1999 event (Figure 5d). This strengthens the climatological wind there, corresponding to favorable emission conditions and therefore resulting in reduced aerosol concentrations (Figure 1a). While there are anomalous southwesterslies/southerlies over north China in the winter of the 2000/2001 event, this led to a weakening of the climatological wind and therefore increased aerosol concentrations in the mature phase of the 2000/2001 event (Figure 1d).

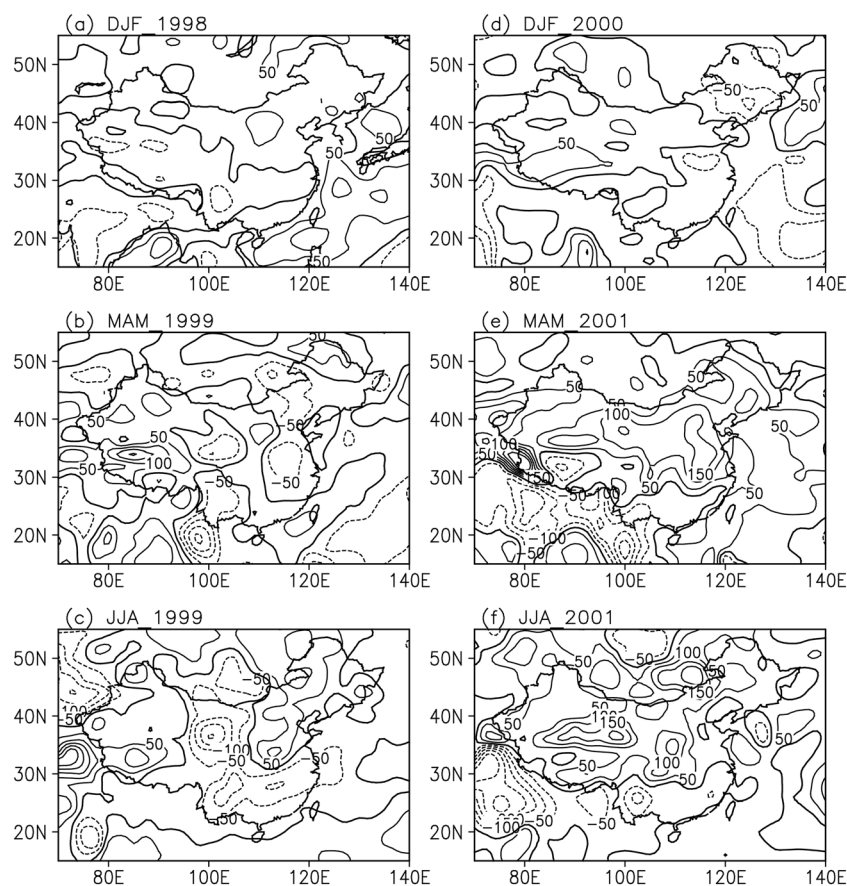


Figure 7. Horizontal distribution of PBLH (m) based on assimilated meteorological data during the La Niña Modoki events of (left) 1998/1999 and (right) 2000/2001.

In the spring of the 1998/1999 decaying phase, the negative SSTA in the central Pacific is weakened and shifted westward. Accordingly, the positive SSTA in the western Pacific is reduced, but not that in the North Pacific (Figure 4b). In response to this SSTA pattern, the anomalous cyclonic flow in the south China coastal regions displays a northeastward shift (Figure 5e). Consequently, eastern China is affected by anomalous southeasterlies/easterlies, indicating input of clean air in this region. For the decaying phase spring of the 2000/2001 event, the warm SSTA in the North Pacific was aligned northeast-southwest, parallel to a reduced cooling in the central Pacific (Figure 4e). Under the influence of this northeast-southwest SSTA pattern in the western Pacific, a belt of strong convergence develops over the eastern coastal regions of China, associated with an anomalous cyclonic circulation in the North Pacific centered around 30°N, 145°E, with anomalous northwesterlies north of 30°N, but northeasterlies south of 30°N over most of eastern China (Figure 5h). That is, eastern China is controlled by an anomalous anticyclonic circulation. The anomalous northerlies in north China strengthen the climatological wind (Figure 5b) and therefore favor aerosol diffusion. However, the anomalous convergence around 30°N indicates that more pollutant is concentrated along this latitude. Moreover, there are evident negative wet deposition anomalies during this season (Figure 8d). The above results suggest a combined role of circulation and wet deposition that contributes to the anomalous aerosol concentrations during the decaying spring of the 2000/2001 event over eastern China.

During the summer of the 1998/1999 decaying phase, the extent of cool SSTA in the central Pacific is continuously reduced and shifts eastward, while the warm SSTA shifts farther north (Figure 4c). The anomalous cyclonic circulation moves northeastward, with the main body over eastern China (Figure 5f). This circulation indicates that convergence is enhanced over eastern China, corresponding to increased aerosol concentrations during the decaying phase summer of the 1998/1999 event (Figure 1c). In the 2000/2001 event, the cool SSTA in the central Pacific almost disappears, whereas the warm SSTA along the eastern China coastal regions remains (Figure 4f). As a response to this SSTA pattern, anomalous convergence is induced along

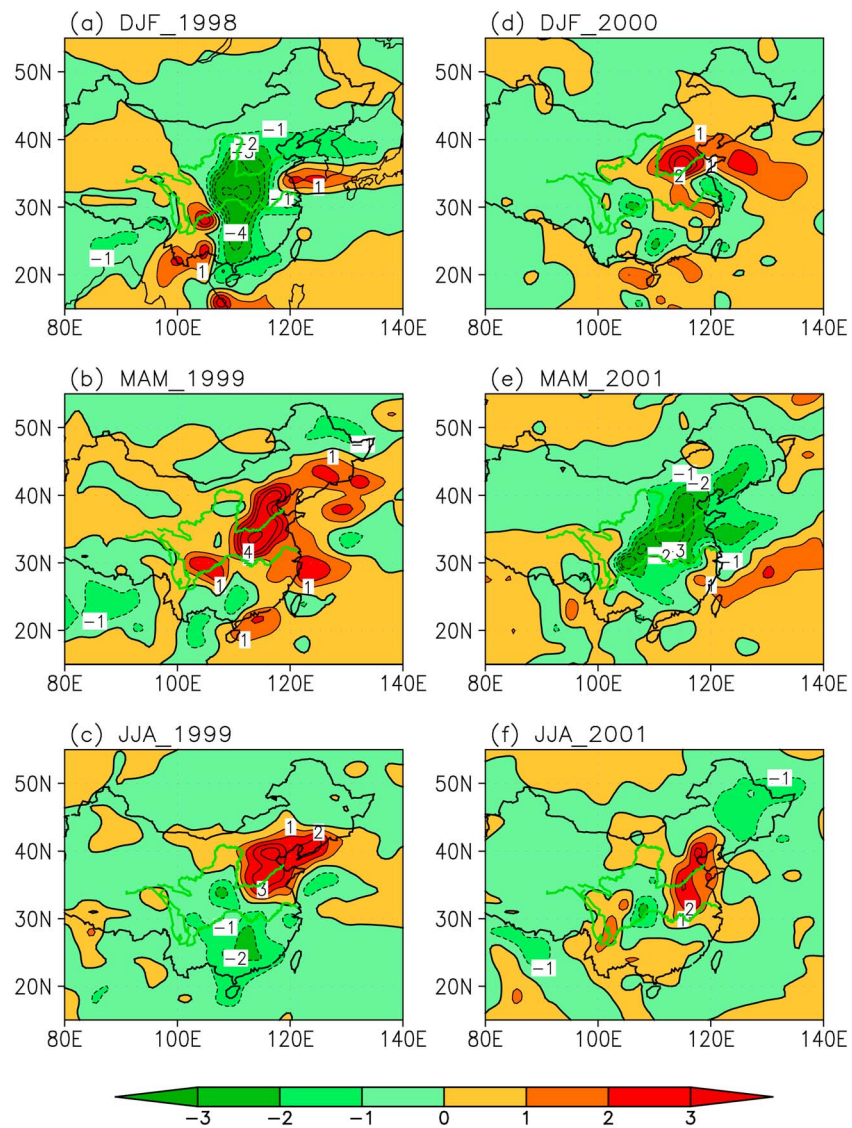


Figure 8. As in Figure 7, but for the distribution of vertically integrated wet deposition flux anomalies during the La Niña Modoki events of (left) 1998/1999 and (right) 2000/2001.

the eastern China coastal regions. Consequently, south China is influenced by easterly/northeasterly anomalies (Figure 5i), leading to increased aerosol concentrations. However, north China is controlled by anomalous southerlies/southeasterlies (Figure 5i), a circulation that reduces aerosol emissions (Figure 1f).

Further, the vertical integral of aerosol concentrations fluxes at the four boundaries over eastern China is examined (Figure 6). It is found that the anomalies in the decaying spring and summer of the 1998/1999 event are largely due to the zonal transport. However, the dipole pattern during the mature winter of the 1998/1999 event is a combined effect of zonal and meridional transports. For the 2000/2001 event, the anomalous aerosol concentrations during the mature and decaying seasons are mainly due to the zonal transport, with the meridional transport much smaller than the zonal transport.

We now examine the effect of atmospheric stability (i.e., planetary boundary layer height, PBLH) on aerosol concentrations (Figure 7). We see that the anomalous distribution of PBLH is consistent with the distribution of aerosol concentrations, except in the mature winter of the 2000/2001 event. For example, positive anomalies in PBLH are observed in north China and negative PBLH anomalies in south China during the spring and summer of the 2000/2001 event, accompanied by enhanced aerosol concentrations over south China but decreased aerosol concentrations over north China. That is, the variation of PBLH shows a negative

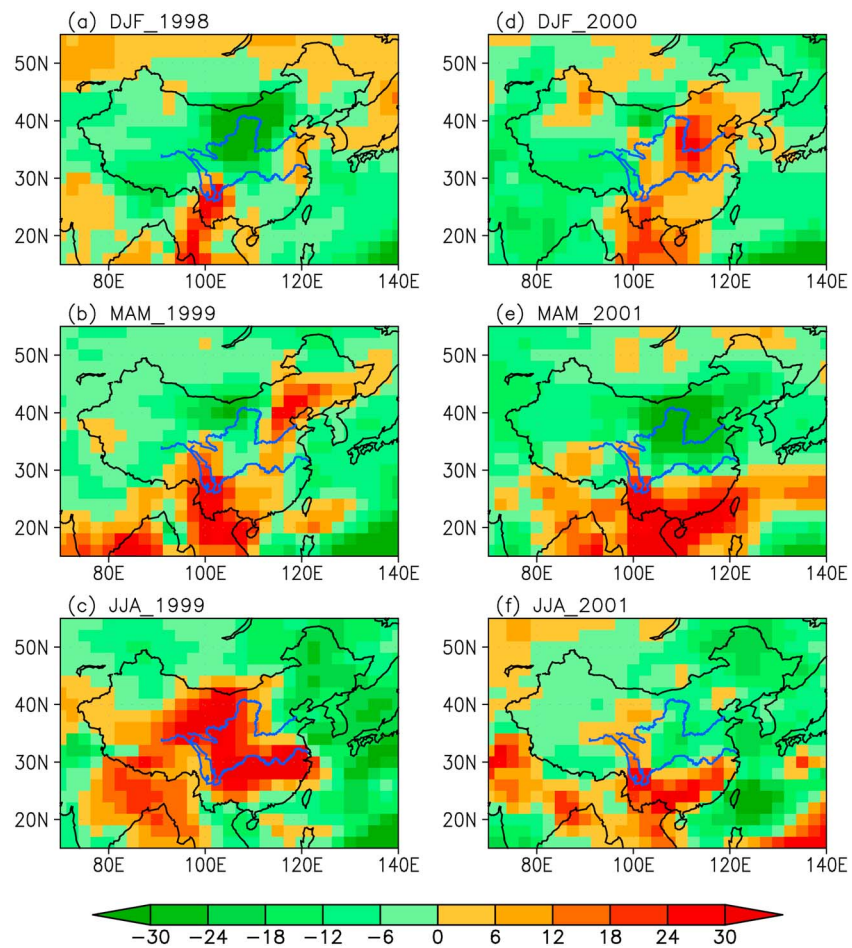


Figure 9. As in Figure 1, but for the spatial distribution of the surface-layer $PM_{2.5}$ RPAD in the sensitivity run when the wet deposition is turned off.

relationship with the aerosol concentrations, which is consistent with previous studies [Dawson *et al.*, 2007; Jacob and Winner, 2009]. Note, however, that the role of PBLH on the aerosol concentrations shows certain differences and varies with region and season.

Besides the possible influences from the circulation, wet deposition may exert a certain role in determining the distribution of aerosol concentrations [Wu, 2014]. We further examined the wet deposition anomalies associated with the two La Niña Modoki events. It is found that the pattern of wet deposition anomalies is not consistent with those observed in the aerosol concentrations (Figure 8). Negative anomalies of wet deposition are clearly seen during the mature phase of the 1998/1999 event (Figure 8a) and in the decaying spring of the 2000/2001 event over eastern China (Figure 8e). The anomalies in the wet deposition are associated with positive aerosol concentration anomalies. This is consistent with the observed anomalies over south China but not for north China. The increased wet deposition over north China in the mature phase of the 2000/2001 event (Figure 8b) and decaying phase summer of the 1998/1999 event (Figure 8c) is in conflict with the observed enhanced aerosol concentrations there.

A sensitivity simulation was performed to quantify the potential influence of wet deposition on the aerosol concentrations by turning off the wet deposition (Figure 9). It is found that the distributions of aerosol concentration anomalies are similar to those shown in Figure 1 during the decaying phases for the 2000/2001 event and the decaying summer of the 1998/1999 event in both north and south China. Similar distributions are also found in north China during the mature phases of both events and the decaying spring of the 1998/1999 event, but not in south China. This result further confirms that the rainfall plays a limited role in determining the anomalous aerosol concentrations during the two La Niña Modoki events. This result

agrees with Wu [2014] and Feng *et al.* [2016a] regarding the seasonal and regional variations of the effect of rainfall on aerosol concentrations.

5. Summary and Discussion

Using simulations with the GEOS-Chem model, we explored the potential influences of the two La Niña Modoki events on the aerosol concentrations over eastern China during their lifespan. In particular, the possible impacts of strong and moderate La Niña Modoki events on aerosol concentrations are discussed. For the strong La Niña Modoki event of 1998/1999, an obvious meridional dipole pattern of aerosol anomalies is observed in the mature phase over eastern China. However, an opposite dipole pattern is seen in the mature phase of the 2000/2001 event, implying totally opposite impacts of the strong and moderate La Niña Modoki events on the distribution of aerosol concentrations over eastern China. In the decaying spring of the 1998/1999 event, there are generally reductions in aerosol concentrations over eastern China, whereas in the decaying summer, aerosol concentrations increased. During the decaying spring and summer of the moderate La Niña Modoki event of 2000/2001, the dipole pattern in eastern China is opposite to that of the mature phase. These results suggest that the impacts of the strong and moderate La Niña Modoki events on aerosol concentrations over eastern China differ over their lifespan. Note that the stronger northerly anomalies during the decaying spring of 2000/2001 (Figure 5h) would transport more dust aerosols to north China in reality. In fact, the aerosol concentrations in the present work mainly refer to anthropogenic aerosols. However, the dust aerosols contribute a considerable proportion of the total aerosol concentrations, especially in the boreal spring over north China. At present, due to the limitations of the observations and model capability, we could not give a quantitative assessment of the influences of La Niña events on natural aerosols. A recent work reported that the variation of EASM has a significant impact on the dust concentrations over eastern China and highlighted the role of atmospheric circulation for natural aerosols [Lou *et al.*, 2016]. On this point, the possible influences of ENSO on the natural aerosols deserve further consideration.

Unlike the strong La Niña Modoki event of 1998/1999, the amplitude of the cooling SSTA in the central Pacific of the moderate La Niña Modoki event of 2000/2001 is smaller and shows rapid weakening during its development. During the strong La Niña Modoki event 1998/1999, the anomalous cyclonic circulation over the south China coastal regions plays an essential role in determining the aerosol concentrations during its lifespan. The location of this anomalous cyclonic circulation results in a different aerosol distribution. In contrast, the SSTA in the North Pacific plays an important role in the moderate La Niña Modoki event of 2000/2001. This SSTA related convergence would impact the circulation over eastern China and thus influence the aerosol concentrations there.

As mentioned above, the SSTA in the North Pacific plays an important role in the La Niña Modoki air-sea interactions and in forming the related circulation anomalies. The possible role and the formation of the anomalous SST in the North Pacific have been discussed in Ding *et al.* [2015a, 2015b]. It is reported that the SST variations in the North Pacific (i.e., its second dominant mode) are linked to the development of ENSO Modoki events and could act as an ocean bridge to influence ENSO Modoki. On this point, it is worth examining further the link between the North Pacific SST and ENSO Modoki, considering that there are large differences in SSTA in the North Pacific between the strong and moderate events. It is of interest to further investigate whether the leading signal in the North Pacific shows any differences if the events are classified into strong and moderate La Niña Modoki events.

We further extended the study period to cover 1948–2010. There are two strong and four moderate La Niña Modoki events during this period, i.e., 1988/1989 and 1998/1999 for the strong La Niña Modoki events and 1973/1974, 1975/1976, 1983/1984, and 2000/2001 for the moderate events. However, some of the La Niña Modoki events coincided with anomalous EAWM events. For example, the winter of 1983/1984 corresponds to a strong EAWM event and the winter of 1973/1974 corresponds to a weak EAWM event [Jhun and Lee, 2004]. Thus, it is difficult to separate the individual influences of La Niña Modoki and the EAWM on the anomalous circulation over eastern China. In addition, since the influences of warm and cold ENSO Modoki events on regional climate are asymmetric [Cai and Cowan, 2009; Feng and Li, 2011], we could not use correlation or regression methods to further examine the influences of La Niña Modoki on aerosol concentrations. Besides, as shown above, the impacts of strong and moderate La Niña Modoki events on aerosol concentrations over eastern China are different or opposite, so a composite of all the events may offset or

emphasize the relative role of certain events. Therefore, long-term and high-quality aerosol observations are needed to further examine the climatic effects of La Niña Modoki events on aerosol concentrations in the future.

Independent of this study, *Feng et al.* [2016a] reported that the El Niño Modoki event has a large impact on aerosol concentrations over southern China. Comparing with their results, we see that the impacts of the corresponding warm events on aerosol concentrations over south China are not linearly opposite. A clear asymmetric relationship is seen for both the strong and moderate La Niña Modoki events. Although only two events are explored in this study, the results reveal that the influence of the La Niña Modoki on aerosol concentrations cannot be ignored; the impact varies with the development of the event and shows a large dependence on the intensity of the event itself. Due to the complexity of the ENSO Modoki issue, long-term data sets are needed to further clarify the roles that ENSO Modoki events play in influencing aerosol concentrations and to assess the relative roles of ENSO and the monsoons in affecting the distribution of aerosol concentration anomalies. We will further clarify this issue in our future work by employing long period data sets.

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