Manuscript # ACP-2015-804

We thank the reviewers for their comments that are very helpful for improving the quality of the manuscript. We have revised the manuscript carefully, as described in our point-to-point responses to the reviewers' comments.

Response to Reviewer #1

General comments.

I appreciate the authors' efforts in improving the manuscript. But I still show some concerns about the conclusions.

1. The authors use PNA+ (PNA-) to represent the months with top (bottom) 25% PNA values. This is misleading for many readers who don't read this paper very carefully or just go through the figures very quickly. We usually think PNA+ are those months with positive (above the median value) values. The authors should use a different symbol or specify their definition carefully in the figures and main text.

Response:

Thanks for the suggestions. The method of composite analysis to examine the differences in aerosol concentrations between the top and bottom values of the index of a circulation system was commonly used by previous studies (Eckhardt et al., 2003; Jerez et al., 2013; Zhu et al., 2012). To avoid confusion, we have added the definition of PNA+ (PNA–) in the abstract and figure captions to state clearly that the PNA positive months are the 25% of the months with the highest positive PNAI values and the negative PNA months (PNA–) are 25% of the months with the highest negative PNAI values.

2. The authors concludes that 'the PNA-induced variation in planetary boundary layer height was found to be the most dominant meteorological factor that influenced the concentrations of PM2.5, sulfate, ammonium, organic carbon, and black carbon, and the PNA-induced variation in temperature was the most important parameter that influenced nitrate aerosol'. All of these results just come from a CTM study without using the observations. In order to show PBL is the major driving factor, I have suggested that the authors should do a sensitivity test by fixing the PBL, but they think their statistical methods are very robust and don't do this test. In Figure 7, the PBL height decreases only 5% in the eastern US. I am skeptical if this will still pass the significance test if they use a p-value 0.05 instead of 0.10. Are these results still true using a different PBL scheme or a different reanalysis met field? Also they don't explain why PBL will decrease under PNA+ months. I think the authors should address the following questions before this paper is accepted. Response:

Following the Reviewer's suggestion, we have done a significance test with the p-value of 0.05. Fig. A shows the the absolute and relative differences in PBLH between PNA+ and PNA– months, with the p-value of 0.05. The differences are similar to Fig. 7 (with the p-value of 0.10) in the manuscript. In Fig. A, the region with 5% difference in PBLH passes the significant test with the p-value of 0.05 in the eastern US.

The GEOS-Chem can use two PBL mixing schemes, the full PBL mixing (TURBDAY) scheme and the Non-local PBL mixing scheme (VDIFF). The TURBDAY is used in our work and the VDIFF scheme can only be used with the GEOS-5 meteorology (http://wiki.seas.harvard.edu/geos-chem/index.php/Boundary_layer_mixing). However, the GEOS-5 version 5.1.0 and 5.2.0 meteorological fields are available for years 2003 to 2008 and years of 2008 to 2013 respectively (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-5), which are not suitable for our study that needs time series on decadal time scales.

Previous studies examined the impacts of PNA on temperature, humidity, and precipitation as described in Sect. 5.2 of our manuscript, but few studies investigated the impacts of PNA on PBL in the U.S.. We have added sentences to explain the possible mechanism for PBLH differences between PNA+ and PNA– phases in Sect. 5.2: "The changes of PBLH were possibly associated with the frequent outbreak of cold air in the U.S. in PNA+ phases (Leathers et al., 1991; Archambault et al., 2010), which induced cool air at the surface and clouds over the intermountain and the eastern Midwest regions (Sheridan, 2003)." The understanding of the impact of PNA on PBLH requires future studies.

Since the changes in PBLH are associated with the changes in other meteorological parameters, we have revised our conclusion in abstract as "Based on the GEOS-Chem simulation, the pattern correlation coefficients were calculated to show the impacts of PNA-induced variations in meteorological fields on aerosol concentrations. The PNA phase was found to influence sulfate concentrations mainly through changes in planetary boundary layer height (PBLH), precipitation (PR), and temperature, to influence nitrate concentrations mainly through changes in temperature, and to influence concentrations of ammonium, organic carbon, and black carbon mainly through changes in PR and PBLH."



Fig. A. Similar to Fig. 7 in the manuscript except that the white spaces indicate the areas that did not pass the two-tail student-t test with 95% significance level.

First, this conclusion is made without using the observations. If we compare the PNA-driven differences in PM2.5 between observation (Figure 3) and GEOS-Chem (Figure 5), we can find many big discrepancies. For example, little effect of PNA on PM2.5 is found in California in GEOS-Chem. As a result, there is no discussion about California, a very important state for the US, in Section 5.2. To our knowledge, present CTMs have many problems in simulating the aerosol variability. As such, the analysis only based on CTM results can hardly contribute to our understanding of the effects of PNA on PM2.5. I think the authors should re-do the analysis in Table 4 using observations. They can use PBL from GEOS4 since there are no PBL observations.

Response:

In the manuscript, we do not calculate pattern correlation coefficients by using observed aerosol concentrations, because observations are influenced by complex variations in local emissions, which would impact the analysis of the relationship between aerosol concentrations and meteorological fields.

Following the Reviewer's suggestion, we have tried to calculate pattern correlation coefficients (PCC) between the composite differences in observed aerosol concentrations and the corresponding composite differences in meteorological parameters in the U.S. (see Table A below). Table A shows that the PBLH had largest PCC values for PM_{2.5}, NH⁺₄, OC and BC, which agree with the results in Table 4. For PM_{2.5}, precipitation (PR), wind speed (WS) and PBLH were important. The PNA-induced variations in temperature had a strong negative correlation (PCC=-0.73) with the PNA-induced differences in nitrate concentrations. The PCC values for observed SO_4^{2-} are found to be insignificant in Table A, because the impact of PNA on SO²⁻ concentrations was not significant in the U.S. on the basis of EPA-AQS data (Table 1). There are also some differences between Table 4 (on the basis of GEOS-Chem simulation with fixed anthropogenic emissions during the simulation for years of 1986–2006) and Table A (on the basis of observed concentrations) for the following two reasons. First, the observations were influenced by complex variations in local emissions. Second, the time series of observed concentrations are much shorter than the GEOS-Chem model results for years of 1986-2006.

Table A. The pattern correlation coefficients (PCC) between the composite differences in EPA-AQS aerosol concentrations and the corresponding composite differences in meteorological parameters in the western U.S.. The * denotes the correlations that have passed the two-tail t-test with 95% confidence level. Note that T, RH, and surface wind speed are from the NCEP

Reanalysis, precipitation is from the Global Precipitation Climatology Center (GPCC) data, and PBLH is from the GEOS-4 data in 1999–2006 for PM_{2.5} and 2000–2006 for other species. The EPA-AQS aerosol concentrations and meteorological variables are regrided into $2^{\circ} \times 2.5^{\circ}$ grids to be consistent with Table 4 in the manuscript. The seasonal cycle and trend in the observed aerosol concentrations are removed.

	PM _{2.5}	SO ₄ ²⁻	NO ₃	NH_4^+	OC	BC
Т	-0.13	0.16	-0.73*	-0.31*	-0.37*	-0.46*
PR	-0.32*	0.04	-0.02	-0.20*	-0.19*	-0.21*
RH	0.11	-0.46*	0.24*	0.31*	0.30*	0.32*
WS	-0.37*	0.13	-0.12	-0.36*	-0.14*	-0.09
PBLH	-0.73*	0.47*	-0.33*	-0.70*	-0.60*	-0.70*

Second, they think the PBL height is very important in explaining the variability in PM2.5, which may not be true in some regions. One issue is that this paper never explains why the PBL will change. Any analysis on this topic will be very useful to this community. Figure 7 shows that the PBL in the Midwest changes fewer than 25 meters (<5%) in the Midwest. However, the PNA-driven changes in PM2.5 are 20%-40% in observations (Figure 3) and 10%-20% in GEOS-Chem in this region. According to these numbers, it seems that the PBL shouldn't be an important factor in explaining the PNA-driven changes in the Midwest. For the intermountain west, I agree with the authors' conclusion that PBL should be an important factor according to Figure 7, but this conclusion is subject to large uncertainties due to the complex topography there. The authors should make this clear in the conclusion and discussion. Response:

Considering that the changes in PBLH are associated with the changes in other meteorological parameters, we have removed our strong conclusion about PBLH in the manuscript. For example, we have revised our conclusion in abstract as "Based on the GEOS-Chem simulation, the pattern correlation coefficients were calculated to show the impacts of PNA-induced variations in meteorological fields on aerosol concentrations. The PNA phase was found to influence sulfate concentrations mainly through changes in planetary boundary layer height (PBLH), precipitation (PR), and temperature, to influence nitrate concentrations mainly through changes in temperature, and to influence concentrations of ammonium, organic carbon, and black carbon mainly through changes in PR and PBLH."

Third, they think 'PNA-induced variation in temperature was the most important parameter that influenced nitrate aerosol'. This conclusion should be correct due to our present knowledge in atmospheric chemistry. But the observed and

simulated NO3 changes in Figure 3 and Figure 5 are not quite different in spatial distribution. So the authors need to clarify this. Response:

We have added a sentence to clarify the differences between the observed and simulated NO_3^- changes the Sect. 4.2: "The model underestimates the differences in NO_3^- in California and the southeastern U.S., because the model does not capture well the temporal variations of NO_3^- in these two regions (Fig. S4)."

3. Section 5.1 raises many questions. First, why the box in Figure 6b doesn't include the whole California?

Response:

Because the whole of the U. S. is not a regular rectangle, we choose the box to include most parts of the continental U. S. and try to include small ocean areas, considering that we are focused on aerosol concentrations in the continental U. S.. Our conclusion still holds if we shift the west boundary to further west (see our responses to your next two comments).

Second, why the mass fluxes are integrated from surface to 100 hPa? The top layer (100 Pas) is already in the stratosphere, which shouldn't have big influence on the surface.

Response:

Following the Reviewer' suggestion, we have revised the results in Table 3 to calculate fluxes from the surface to 250 hPa. The relative change in net mass flux is 13.3 kg s⁻¹ (9.9%) in the revised Table 3, similar to that of 14.0 kg s⁻¹ (11.4%) in our previous version when we integrated from surface to 100 hPa. Therefore, the conclusion from the section is not affected.

Third, numbers in Table 3 will change a lot if the location of the boundary is shifted. For example, if the west boundary is in the ocean, the transport via west boundary should be close to 0.

Response:

We have tried to shift the west boundary to 125° W to include the whole California in the box. The selection of box does not compromise our conclusion; the difference in net mass flux between PNA+ and PNA– is 13.3 kg s^{-1} in Table 3 of our manuscript, and it is 12.6 kg s^{-1} in Table B below.

Table B. Same as Table 3 in the manuscript except that selected box includes the whole California (75–125°W, 28–49°N, from the surface to 250 hPa).

	Boundaries and total	Mass Flux
	West	86.2
	East	233.9
PNA+	South	8.3
	North	-15.9
	Net flux	-123.4

	West	98.4
	East	247.4
PNA-	South	24.7
	North	11.8
	Net flux	-136.0
	West	-12.2
Diff	East	–13.5
DIII. (DNA + minus DNA)	South	-16.4
(FINAT IIIIIUS FINA-)	North	-27.7
	Net flux	12.6

Fourth, the authors should explain why the underestimate in GEOS-Chem over west US won't compromise this conclusion in more details. The underestimate should change the transport via west boundary a lot. Please consider if this section is needed for this paper.

Response:

We have added the following sentence to the end of Sec. 5.1: "Note that because the GEOS-Chem model underestimates $PM_{2.5}$ concentrations in the western U.S. (Fig. 4b), the net outflow flux from the selected box might have been underestimated, but this should not compromise our conclusions about the relative differences in net flux between PNA+ and PNA– phases."

Minor comments.

Figures. If detrended PM2.5 concentrations are used, please clarify in the captions. Does Figure S4 use the detrended data?

Response:

We have clarified that we use detrended data in the captions of Fig. 3, S2, S3 and S4: "The seasonal cycle and trend in observed aerosol concentrations are removed as described in Sect. 2.1".

Introduction. Please be more accurate when citing previous results. For example, the exact number of climate-driven $PM_{2.5}$ changes is ±0.1-1 µgm⁻³ (P4 L81). Please also check other sentences. Response:

The climate-driven changes in $PM_{2.5}$ were ±0.1–1 µg m⁻³ in the study of Jacob and winner (2009), which referred to the regions of North America and Europe (see Sect. 8 of Jacob and winner (2009)). Here we are focused on the U.S.. The number in the sentence "Future climate change was also simulated to influence aerosol levels over the U.S. by about 1 µg m⁻³ (Jacob and Winner 2009)" is obtained from Table 3 of Jacob and winner (2009). To be more accurate, we have revised the sentence as "Future climate change was also simulated to influence aerosol levels over the U.S. by -1 to +1 µg m⁻³ (Jacob and Winner 2009)". We have also checked and corrected other sentences.

P180. Please clarify the method you used here instead of asking readers to jump over to the later section.

Response:

We have changed "(see Sect. 2.3 below)" to "(the aerosol concentrations without seasonal cycle are defined as $C'_{i,j} = C_{i,j} - \frac{1}{n} \sum_{i=1}^{n} C_{i,j}$, where $C_{i,j}$ is the aerosol concentrations in month *j* of year *i*, *n* is the number of years examined)".

P187. When you fit the regression model, do you use data only in the winter or the whole year? Be more accurate here. Response:

We have clarified this as "..., the long-term linear trend in concentrations is identified by the least-square fit and then removed from the wintertime observed concentrations for each site."

P215. What is the exact height of PBL? The readers don't know these details of GEOS-Chem vertical resolution.

Response:

We have clarified here that "The average PBL height in wintertime in the U. S. is about 480 m and occupies the lowest 3–6 vertical model layers."

P229. PNA phases are only part of meteorological parameters here. Response:

We have deleted "(PNA phases)" here.

P289-290. The numbers here are misleading. Are these numbers the average change of all sites in this region? Seems not.

Response:

We have clarified this as "The maximum enhancement of $PM_{2.5}$ reached 7–9 µg m⁻³ (or 40–80%) in California,..."

P309-310. Change 'were' to 'are'. Please fix many other similar problems in the whole text.

Response:

Changed.

P347. It seems the PNA can only explain a very small fraction (<15%) of the variability. Is PNA important for aerosol? Please give some discussion here. Response:

Such FTVEP values are not so small because the value of 15% denotes a correlation coefficient (CC) of 0.39, passing the t-test with the confidence of 99.9%. Many previous studies reported large correlations with similar CC and FTVEP values (see Table C below).

We have the following sentences on the importance of PNA in the text "For $PM_{2.5}$, SO_4^{2-} , NO_3^{-} and NH_4^{+} aerosols, FTVEP values were high over and near

the eastern Midwest, where the PNA teleconnection explained up to 50%, 40%, 50%, and 40% of temporal variances of surface concentrations of these aerosol species, respectively.

Table	С.	Previous	studies	that	reported	large	correlations	with	similar
correla	ation	s coefficie	nts and F	TVE	> .				

References	Relationships	CC	FTVEP
Gong et al. (2001)	Arctic Oscillation vs. the	0.34	12%
	temperature in the eastern China		
Gong et al. (2006)	Ratio of dust inflow to North	0.29	8%
	America and outflow over North		
	Pacific vs. the western Pacific		
	pattern (WP)		
Müller et al. (2006)	Nino 3.4 Index (denoting the	0.26-0.42	7–17%
	ENSO) vs. PNA		
Xiao et al. (2014)	detrended Haze day in China vs.	0.47	22%
	the Atlantic multidecadal oscillation		
	(AMO)		
Zhou et al., (2015)	PM ₁₀ concentration in Shanghai ir	n <u>–</u> 0.38	14%
	winter vs. surface wind speed		

P349. How many sites show high FTVEP values? Please provide the fraction number.

Response:

We have added a sentence to provide the fractions passing the t-test of correlation coefficients for each species: "About 40%, 30%, 47%, 33%, 33% and 34% of sites pass the two-tail t-test with 90% confidence for $PM_{2.5}$, SO_4^{2-} , NO_3^- , NH_4^+ , OC and BC, respectively."

P469-471. Obviously the model doesn't capture the difference in California. Please be more accurate here.

Response:

We have revised the sentence as "simulated geographical patterns of the differences in $PM_{2.5}$ and each aerosol species between the PNA+ and PNA– months were similar to those seen in observations in most areas of the U.S. (except for California)."

References:

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Response to Reviewer #3

1. The correlations are found between PNA-driven meteorological variables (e.g., T, PR, RH, ...) and many types of aerosols (e.g., SO4, NO3, NH4,...) with pattern correlation coefficients. These correlations provide some statistical

implications on the potential mechanisms of PNA influence on aerosol concentrations. However, the manuscript in many places reads like those correlations suggest a causal relationship between PNA phases and aerosol loadings. I recommend rephrasing the conclusion by explicitly noting that the results are merely correlations. More evidence is needed to reach a solid conclusion.

Response:

Following the Reviewer's suggestion, we have revised our conclusions in abstract and conclusion section:

In the abstract: "Based on the GEOS-Chem simulation, the pattern correlation coefficients were calculated to show the impacts of PNA-induced variations in meteorological fields on aerosol concentrations. The PNA phase was found to influence sulfate concentrations mainly through changes in planetary boundary layer height (PBLH), precipitation (PR), and temperature, to influence nitrate concentrations mainly through changes in temperature, and to influence concentrations of ammonium, organic carbon, and black carbon mainly through changes in PR and PBLH."

In conclusion section: "It should be noted that, the PCC values only statistically present the relationship between meteorological parameters and aerosol concentrations in the U.S.. More in-depth understanding of the impact of PNA phase on aerosol concentrations should be carried out on the basis of physical and chemical processes."

2. The GEOS-Chem OH results are documented in the author response, but not in the main text. It would be helpful that this information be included in the revised version, as it provides information about the aerosol sinks in the simulation.

Response:

Following the suggestion, we have added a sentence into Sect. 2 to provide the information on OH: "The OH concentrations in GEOS-Chem were examined by previous studies (Holmes et al., 2013; Wu et al., 2007) by calculating the methane lifetime, which agreed closely with the lifetime of 11.2±1.3 yr constrained by methyl chloroform observations by Prather et al. (2012). Previous studies also showed that aerosol concentrations were not so sensitive to OH concentrations in the GEOS-Chem simulations (Heald et al., 2012)."

References:

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1	The Impact of Monthly Variation of the Pacific-North America (PNA) Teleconnection
2	Pattern on Wintertime Surface-layer Aerosol Concentrations in the United States
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26 Abstract

27 The Pacific-North America teleconnection (PNA) is the leading general circulation pattern in the troposphere over the region of North Pacific to North America during 28 29 wintertime. This study examined the impacts of monthly variation of the PNA phase 30 (positive or negative phase) on wintertime surface-layer aerosol concentrations in the 31 U.S. by analyzing observations during 1999–2013 from the Air Quality System of the 32 Environmental Protection Agency (EPA-AQS) and the model results for 1986–2006 from the global three-dimensional Goddard Earth Observing System (GEOS) 33 34 chemical transport model (GEOS-Chem). The composite analyses on the EPA-AQS 35 observations over 1999–2013 showed that the average concentrations of PM_{2.5}, 36 sulfate, nitrate, ammonium, organic carbon, and black carbon aerosols over the U.S. 37 were higher in the PNA positive phases (25% of the winter months examined, and this 38 fraction of months had the highest positive PNA index values) than in the PNA 39 negative phases (25% of the winter months examined, and this fraction of months had the highest negative PNA index values) by 1.0 μ g m⁻³ (8.7%), 0.01 μ g m⁻³ (0.5%), 40 0.3 μ g m⁻³ (29.1%), 0.1 μ g m⁻³ (11.9%), 0.6 μ g m⁻³ (13.5%), and 0.2 μ g m⁻³ (27.8%), 41 42 respectively. The simulated geographical patterns of the differences in concentrations 43 of all aerosol species between the PNA positive and negative phases were similar to 44 observations. Based on the GEOS-Chem simulation, the pattern correlation 45 coefficients were calculated to show the impacts of PNA-induced variations in 46 meteorological fields on aerosol concentrations. The PNA phase was found to 47 influence sulfate concentrations mainly through changes in planetary boundary layer height (PBLH), precipitation (PR), and temperature, to influence nitrate 48 49 concentrations mainly through changes in temperature, and to influence concentrations of ammonium, organic carbon, and black carbon mainly through 50

- 51 changes in PR and PBLH. Results from this work have important implications for
- 52 understanding and prediction of air quality in the United States.

53 **1** Introduction

Aerosols are the major air pollutants that have adverse effects on human health, reduce atmospheric visibility, and influence climate through aerosol-radiation and aerosol-cloud interactions (IPCC, 2013). Aerosol concentrations are high over the industrialized regions such as the U.S., Europe, and East Asia, which are driven by emissions of aerosols and aerosol precursors (Dutkiewicz et al. 2000; Vestreng et al. 2007; Hand et al. 2012a; Mijling et al. 2013) and regional meteorological conditions.

60 Previous studies have shown that aerosol concentrations are very sensitive to 61 meteorological parameters (Aw and Kleeman 2003; Wise and Comrie 2005; Dawson 62 et al. 2007; Kleeman 2008; Jacob and Winner 2009; Tai et al. 2010; Tai et al. 2012a; 63 Allen et al. 2015; Markakis et al. 2015; Megaritis et al. 2014; Porter et al. 2015). Aw 64 and Kleeman (2003) examined the sensitivity of PM_{2.5} (aerosol particles which 65 diameter $\leq 2.5 \ \mu$ m) concentration to temperature by performing sensitivity studies in the California Institute of Technology/UC Davis (CIT/UCD) air quality model. A 66 67 cross-board increase in temperature by 5 K in Southern California on September 25, 1996, led to decreases in peak PM_{2.5} concentrations by up to 30.7 μ g m⁻³ (~30%). 68 Wise and Comrie (2005) reported, by statistical analyses of observational datasets 69 obtained from Air Quality System of U.S. Environmental Protection Agency 70 71 (EPA-AQS), that the variations in meteorological parameters accounted for 20-50% 72 of the variability in aerosol levels over 1990-2003 in five metropolitan areas in the 73 southwestern U.S.. They found that aerosols in these five cities were most sensitive 74 to relative humidity. Dawson et al. (2007) found, by sensitivity studies in the 75 Particulate Matter Comprehensive Air Quality Model with extensions (PMCAMx), that PM_{2.5} concentrations in summer had a small sensitivity to temperature increases (-16 76 ng m^{-3} K⁻¹ on average) because the increases in sulfate offset the decreases in 77

78 nitrate and organics, while PM_{2.5} concentrations in winter decreased significantly with temperature (-170 ng m⁻³ K⁻¹ on average) because the increases in temperature led 79 to large reductions in nitrate and organics. Dawson et al. (2007) also showed that 80 81 PM_{2.5} concentrations increased with humidity in both winter and summer. Jacob and 82 Winner (2009) summarized by literature review that the regional stagnation, mixing depth, and precipitation are the most important meteorological parameters that 83 84 influence surface-layer aerosol concentrations. Future climate change was also simulated to influence aerosol levels over the U.S. by -1 to $+1 \ \mu g \ m^{-3}$ (Jacob and 85 86 Winner 2009), as a result of the climate-induced changes in atmospheric oxidants, 87 transport, deposition, and the shift of gas-particle equilibria (Liao et al. 2006; Unger et al. 2006; Bauer et al. 2007; Jacob and Winner 2009; Pye et al. 2009; Lam et al. 2011; 88 89 Day and Pandis 2011; Juda-Rezler et al. 2012; Tai et al. 2012b).

90 Previous studies have also reported that the changes in atmospheric circulation 91 pattern and climate systems, such as the East Asian Summer Monsoon (EASM), 92 North Atlantic Oscillation (NAO), El Nino-South Oscillation (ENSO), Atlantic 93 Multidecadal Oscillation (AMO), and Arctic sea ice (ASI), can modulate distributions 94 and concentrations of aerosols (Moulin et al. 1997; Singh and Palazoglu 2012; Zhu et 95 al. 2012; Jerez et al. 2013; Liu et al. 2013; Xiao et al. 2014; Wang et al. 2015). Zhu et 96 al. (2012) found, by simulation of aerosol concentrations over years 1986–2006 with 97 the global chemical transport model GEOS-Chem, that the decadal-scale weakening 98 of the EASM led to increases in aerosol concentrations in eastern China, and 99 summertime surface aerosol concentrations in the weakest EASM years were larger 100 than those in the strongest EASM years by approximately 20%. Moulin et al. (1997) 101 showed that the variations in NAO could influence mineral dust aerosol transported to 102 the North Atlantic Ocean and the Mediterranean Sea, since the mean aerosol optical

103 depth (AOD) of dust in summer correlated with the NAO index during 1983–1994 with 104 the correlation coefficients of 0.49 and 0.66 respectively. Jerez et al. (2013) found, by 105 simulations of aerosols for years 1970–1999 with the CHIMERE chemistry transport 106 model driven by the BCMWF ERA40 reanalysis data, that the concentrations of PM₁₀ and $PM_{2.5}$ in European differed by 10 and 20 µg m⁻³, respectively, between the 107 108 positive and negative NAO phases. By using the multi-angle imaging 109 spectroradiometer satellite (MISR) datasets of AOD during 2000-2011, Liu et al. 110 (2013) found a period of 3-4 years in observed summertime AOD over the North 111 China Plain (NCP), and the peak of summertime AOD in NCP occurred four months 112 later after the rapidly transition of El Nino from a warm phase to a cold phase because 113 of the associated cyclone anomaly and maritime inflow over the NCP. Singh and 114 Palazoglu (2012) found correlations between PDO and ENSO and the aerosol 115 exceedance days (defined as the days with PM_{2.5} concentrations larger than the U.S. 116 National Ambient Air Quality Standard) at 6 regions in the U.S. by using the EPA-AQS 117 PM_{2.5} wintertime datasets during 1950–2008.

118 The Pacific-North America teleconnection pattern (PNA) is one of the most 119 recognized, influential climate patterns in the mid-latitudes over the region of North 120 Pacific to North America during wintertime with monthly variations (Wallace and 121 Gutzler 1981; Blackmon et al. 1984; Liang et al. 2005; Athanasiadis and Ambaum 122 2009). The PNA phase is defined by the geopotential height anomalies in the middle 123 troposphere over the vicinity of Hawaii, the south of the Aleutian Islands, the 124 intermountain region of North America, and the Gulf Coast region in the U.S. (Wallace 125 and Gutzler 1981). A positive (negative) PNA phase is characterized by positive 126 (negative) geopotential height anomalies over the vicinity of Hawaii and the 127 northwestern North America, while negative (positive) geopotential height anomalies

over south of the Aleutian Islands and the Gulf Coast region (see Fig. S1 in theauxiliary material).

130 The PNA has large impacts on surface-layer meteorological variables in the U.S. 131 during wintertime. Previous studies have reported strong positive (negative) 132 correlation between PNA and surface ambient temperature in the northwestern 133 (southeastern) U.S. (Leathers et al. 1991; Redmond and Koch 1991; Liu et al. 2015), 134 and negative correlation between PNA and precipitation rate (Leathers et al. 1991; 135 Coleman and Rogers 2003; Ning and Bradley 2014; 2015) and moisture (Coleman 136 and Rogers 2003) in the contiguous Ohio River Valley. These variations in 137 meteorological parameters in the U.S. are associated with the PNA-induced 138 anomalies in jet stream position, activities of cold fronts, and synoptic cyclones 139 (Leathers et al. 1991; Notaro et al. 2006; Myoung and Deng 2009).

140 Several studies have examined the impacts of PNA on aerosols. Gong et al. 141 (2006) studied the interannual variations in the trans-Pacific transport of Asian dust 142 during 1960–2003 by using the Northern Aerosol Regional Climate Model (NARCM). 143 They found a negative correlation (with a correlation coefficient of –0.55) between the 144 PNA and the ratio of dust mass that reached the North American continent to that 145 exported from Asia because of the strong westerly jet in the East Pacific during the 146 negative PNA phases. Di Pierro et al. (2011), by using satellite retrieval of aerosol 147 optical depth (AOD) from Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), 148 identified 11 events of Asian aerosol transport to the Arctic during 2007 to 2009, in 149 which 4 events were associated with the negative PNA phases. These studies, 150 however, were focused on the impact of PNA on the transport of aerosols due to the 151 variations in westerly jet stream and blocking activity. Furthermore, these studies 152 were limited to aerosols in the regions of North America and the Arctic.

153 We examine in this work the impacts of monthly variations in PNA phase on aerosol concentrations in the U.S. during wintertime, by analyses of the observed 154 155 aerosol concentrations during 1999-2013 from EPA-AQS and also by simulations of 156 aerosol concentrations for years 1986-2006 using the global chemical transport 157 model GEOS-Chem. The scientific goals of this work are (1) to quantify the 158 differences in wintertime concentrations of sulfate (SO_4^{2-}) , nitrate (NO_3^{-}) , ammonium 159 (NH_4^+) , black carbon (BC), organic carbon (OC), and PM_{2.5} in the U.S. between 160 different PNA phases, and (2) to understand the roles of PNA-induced variations in 161 meteorology (for example, surface air temperature, wind speed, planetary boundary 162 layer height, precipitation, and relative humidity) in influencing the wintertime aerosol 163 concentrations. The definition of the PNA index, the EPA-AQS observation data used 164 in this work, and the numerical simulation with the GEOS-Chem model are described in Sect. 2. Sections 3 and 4 present the impacts of the PNA on wintertime aerosol 165 166 concentrations in the U.S. obtained from the EPA-AQS observations and the 167 GEOS-Chem simulation, respectively. The mechanisms for the impacts of PNA on 168 aerosols are examined in Sect. 5.

169

170 2 Data, simulation, and methodology

171 2.1 Observed aerosol concentrations

172 Observed concentrations of aerosols are obtained from the Air Quality System of the 173 U.S. Environmental Protection Agency (EPA-AQS, 174 http://www.epa.gov/airquality/airdata/). The EPA-AQS daily $PM_{2.5}$ mass 175 concentrations are available over 1999-2013 at about 1200 sites, and the speciated 176 aerosol concentrations, including those of SO_4^{2-} , NO_3^{-} , NH_4^{+} , BC and OC, are 177 available for 2000–2013 at about 300 sites.

178 The measurements of aerosol concentrations from the EPA-AQS were carried 179 out at various time intervals (for example, with measurements every one, three or six 180 days) at different sites, and there were plenty of missing values at many sites. The 181 observed concentrations are pre-processed following the three steps: (1) For a 182 specific site, the observations are used in our analyses if the site had at least 5 183 months of observations and there were at least 5 observation records within each 184 month. (2) The mean seasonal cycle in aerosol concentrations in the months of November-March is removed (the aerosol concentrations without seasonal cycle are 185 defined as $C'_{i,j} = C_{i,j} - \frac{1}{n} \sum_{i=1}^{n} C_{i,j}$, where $C_{i,j}$ is the aerosol concentrations in month j 186 187 of year *i*, *n* is the number of years examined). Such deseasonality approach was used 188 in previous studies that examined the monthly variations in mineral dust aerosol 189 (Cakmur et al. 2001; Mahowald et al. 2003), the decreasing trends in observed PM_{2.5} 190 concentrations and satellite AOD in the southeastern U.S. over 2000-2009 (Alston et 191 al. 2012), and the monthly variations in global AOD (Li et al. 2013). (3) Since the 192 observed aerosol concentrations exhibited a significant decreasing trend from 1999 193 to present in the U.S. due to the reductions in emissions of aerosols and aerosol 194 precursors (Alston et al. 2012. 195 http://www3.epa.gov/airtrends/agtrends.html#comparison), the long-term linear trend 196 in concentrations is identified by the least-square fit and then removed from the 197 wintertime observed concentrations for each site. The EPA-AQS sites with 198 measurements that meet the criteria described in (1) are shown in Fig. 1.

199 2.2 GEOS-Chem Simulation

We also examine the impacts of PNA on simulated aerosol concentrations in the U.S.
by using the GEOS-Chem model (version 8-2-1, http://acmg.seas.harvard.edu/geos).
The GEOS-Chem model is a global chemical transport model driven by the

203 assimilated meteorological fields from the Goddard Earth Observing System (GEOS) 204 of the NASA Global Modeling and Assimilation Office (GMAO). The version of the 205 model we use has a horizontal resolution of 2° ×2.5° and 30 hybrid sigma-P layers 206 from the surface to 0.01 hPa altitude. The model has a fully coupled simulation of tropospheric O_3 -NO_x-VOC chemistry and aerosols including SO_4^{2-} , NO_3^{-} , NH_4^{+} , BC, 207 208 OC (Park et al. 2003; Park et al. 2004), mineral dust (Fairlie et al. 2007), and sea salt 209 (Alexander et al. 2005). Considering the large uncertainties in chemistry schemes of 210 secondary organic aerosol (SOA), SOA in our simulation is assumed to be the 10% 211 carbon yield of OC from biogenic terpenes (Park et al., 2003) and 2% carbon yield of 212 OC from biogenic isoprene (van Donkelaar et al., 2007; Mu and Liao, 2014). We 213 mainly examine simulated anthropogenic aerosols from the GEOS-Chem simulation, 214 since mineral dust concentrations in winter are very small (Malm et al. 2004; Zhang et 215 al. 2013) and sea salt is not a major aerosol species in the U.S. (Malm et al. 2004).

The model uses the advection scheme of Lin and Rood (1996), the deep convective scheme of Zhang and McFarlane (1995), the shallow convection scheme of Hack (1994), the wet deposition scheme of Liu et al. (2001), and the dry deposition scheme of Wesely (1989) and Wang et al. (1998). The instantaneous vertical mixing in the planetary boundary layer (PBL) is accounted for by the TURBDAY mixing scheme (Bey et al., 2001). The average PBL height in wintertime in the U. S. is about 480 m and occupies the lowest 3–6 vertical model layers.

We simulate aerosols for years of 1986–2006 driven by the GEOS-4 reanalysis data. The years of 1986–2006 are chosen for chemistry-aerosol simulation because these are the years that the GEOS-4 datasets are available. Global anthropogenic emissions are from the Global Emissions Inventory Activity (GEIA) (Park et al. 2004; Park et al. 2006; Zhu et al. 2012; Yang et al. 2015). Anthropogenic emissions over the

228 U.S. are overwritten by the U.S. EPA National Emission Inventory for 1999 (NEI99), 229 which have monthly variations in emissions of precursors including SO₂, NO_x, and 230 NH₃. Monthly biomass burning emissions are taken from the Global Fire Emissions 231 Database version 2 (GFED-2) (Giglio et al. 2006; van der Werf et al. 2006). During 232 the simulation of aerosols for years of 1986-2006, the global anthropogenic and 233 biomass burning emissions of aerosols and aerosol precursors are fixed at year 2005 234 levels, so that the variations in aerosol concentrations are caused by variations in 235 meteorological parameters alone.

Natural emissions of O_3 precursors, including biogenic NMVOCs and NO_x from lighting and soil, are allowed to vary over 1986–2006 following the variations in the GEOS-4 meteorological parameters. Biogenic NMVOC emissions are calculated using the module of Model of Emissions of Gases and Aerosols from Nature (Guenther et al. 2006). Lightning NO_x emissions are described by Sauvage et al. (2007) and Murray et al. (2012). Soil NO_x emissions are calculated using the algorithm proposed by Yienger and Levy (1995).

243 The GEOS-Chem simulation of aerosols in the U.S. have been evaluated 244 extensively by previous studies (Park et al. 2003; Park et al. 2004; Park et al. 2005; 245 Heald et al. 2006; van Donkelaar et al. 2006; Liao et al. 2007; Heald et al. 2008; van 246 Donkelaar et al. 2008; Fu et al. 2009; Drury et al. 2010; Leibensperger et al. 2011; 247 Zhang et al. 2012). These studies have shown that the GEOS-Chem model can 248 capture the magnitudes and distributions of aerosols in the U.S.. The OH 249 concentrations in GEOS-Chem were examined by previous studies (Holmes et al., 250 2013; Wu et al., 2007) by calculating the methane lifetime, which agreed closely with 251 the lifetime of 11.2±1.3 yr constrained by methyl chloroform observations by Prather et 252 al. (2012). Previous studies also showed that aerosol concentrations were not so

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255 **2.3 PNA index**

256 The PNA index (PNAI) is commonly used to quantify the changes in PNA phase 257 (Wallace and Gutzler 1981; Leathers et al. 1991). This study follows the definition of 258 PNAI by Leathers et al. (1991). In order to examine the monthly variations in PNA, the 259 mean seasonal cycle of geopotential height at 700 hPa is removed for the months of 260 November, December, January, February, and March (NDJFM) in the studied years. 261 Such deseasonality approach has been used in the analyses of the growth and decay 262 of PNA phase in NDJFM (Feldstein 2002), the development of NAO (Feldstein 2003), 263 the influence of NAO on precipitation in Europe (Qian et al. 2000), and the variations 264 in Madden–Julian oscillation (Wheeler and Hendon 2004). If we are concerned with 265 the PNAI during *n* years, the monthly PNAI in month *j* (*j* is one of the 5 months of 266 NDJFM) of year *i* is calculated by:

sensitive to OH concentrations in the GEOS-Chem simulations (Heald et al., 2012).

267
$$PNAI = \frac{1}{3} \left[-Z_{i,j}^{*\prime}(47.9^{\circ}N, 170^{\circ}W) + Z_{i,j}^{*\prime}(47.9^{\circ}N, 110^{\circ}W) - Z_{i,j}^{*\prime}(29.7^{\circ}N, 86.3^{\circ}W) \right]$$
 (1)

268 where
$$Z_{i,j}^{*'} = \frac{Z_{i,j}'}{\sqrt{\frac{1}{n \times 5} \sum_{i=1}^{n} \sum_{j=1}^{5} Z_{i,j}'^2}}$$
 and $Z_{i,j}' = Z_{i,j} - \frac{1}{n} \sum_{i=1}^{n} Z_{i,j}$. Therefore, $Z_{i,j}'$ denotes the

removal of seasonal cycle, and $Z_{i,j}^{*'}$ denotes the standardized anomaly of geopotential height at 700 hPa in month *j* of year *i* with seasonal-cycle removed.

271 The PNAI is calculated by using both the National Center of Environmental 272 Prediction-Department of Energy Atmospheric Model Inter-comparison Project 273 (NCEP-2, horizontal resolution 2.5°×2.5° Reanalysis data globally. 274 http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis2.html) for years of 275 1986–2013 (referred to as NCEP2-PNAI) and the GEOS-4 assimilated 276 meteorological data (referred to as GEOS4-PNAI) for 1986–2006 (Fig. 2). Both series

of PNA index show strong monthly variations (Fig. 2), and the GEOS4-PNAI agrees
with NCEP2-PNAI over 1986–2006 with a high correlation coefficient of 0.99,
indicating that the NCEP-2 and GEOS-4 datasets are consistent in representing the
monthly variations of PNAI.

There are n-5 PNAI values for *n* years, since we calculate PNAI for the months of NDJFM of each year. These *n*-5 PNAI values are classified into 3 categories for our composite analyses of aerosol concentrations and meteorological parameters: the positive PNA months (PNA+) that are 25% of the *n*-5 PNAI months with the highest positive PNAI values, the negative PNA months (PNA–) that are 25% of the *n*-5 PNAI months with the highest negative PNAI values, and the rest months that are referred to as the transitional months (Fig. 2).

288

289 **3** Impacts of PNA on observed aerosol concentrations

290 The measurements of PM_{2.5} are available over 1999–2013, in which there were 18 291 PNA+ months and 18 PNA- months as shown in Fig. 2a. Figure 3 shows the 292 differences in observed surface-layer PM_{2.5} concentrations between the PNA+ and 293 PNA- months (concentrations averaged over the 18 PNA+ months minus those 294 averaged over the 18 PNA- months). The uncertainty associated with the differences 295 in aerosol concentrations between PNA+ and PNA- months is represented by the 296 two-tail Student-t test with significance level of 90%. Among 1044 sites with PM_{2.5} 297 concentrations (Fig.1), 42% of which had statistically significant differences in PM_{2.5} 298 between PNA+ and PNA- months. Relative to the PNA- months, PM_{2.5} 299 concentrations in PNA+ months were higher in California, the contiguous Salt Lake 300 (northern Utah), and over and near the eastern Midwest. The maximum enhancement of PM_{2.5} reached 7–9 μ g m⁻³ (or 40–80%) in California, 7–9 μ g m⁻³ (80–100%) 301

around the Salt Lake, $3-5 \ \mu g \ m^{-3}$ (40–80%) over and near the eastern Midwest. At 302 303 sites in North Dakota, Wisconsin, Michigan, Minnesota, Montana, Texas and Maine, the PM_{2.5} concentrations were lower by up to 2 μ g m⁻³ (–10 to –20%) in PNA+ months 304 305 than in PNA- months. As the concentrations are averaged over all sites (including the 306 sites that pass and do not pass the t-test with 90% confidence level) in the U.S., the western U.S. (west of 100°W, Fig.1), and the eastern U.S. (east of 100°W, Fig. 1), 307 $PM_{2.5}$ concentrations were higher by 1.0 µg m⁻³ (8.7%), 1.3 µg m⁻³ (14.3%), and 0.8 308 μ g m⁻³ (7.2%), respectively, in the PNA+ months than in PNA– months (Table 1). 309

310 The measurements of speciated aerosols are available during 2000-2013, in 311 which there were 17 PNA+ and 17 PNA- months (Fig. 2b). Figure 3 also shows the 312 differences in observed surface-layer concentrations of individual aerosol species between the PNA+ and PNA- months. The differences in concentrations of SO_4^{2-} , 313 NO_3^- and NH_4^+ show statistically significant positive values at most sites. Among the 314 355, 343, and 194 sites with measurements of SO_4^{2-} , NO_3^{-} and NH_4^{+} , 30%, 44%, and 315 316 39% of which pass the two-tail t-test with 90% confidence level, respectively. While the absolute differences in concentrations of SO_4^{2-} , NO_3^{-} and NH_4^{+} between PNA+ 317 and PNA– were in the range of 0–1 μ g m⁻³ at most sites, the maximum differences 318 reached 1.5–2.5 μ g m⁻³ (30–50%) for SO₄²⁻ in Pennsylvania, 1.5–2.5 μ g m⁻³ (150– 319 200%) for NO $_3^-$ in Illinois, Indiana and Ohio, and 1.5–2.5 µg m $^{-3}$ (50–70%) for NH $_4^+$ in 320 321 Pennsylvania. Averaged over the sites with measurements, the absolute differences in concentrations of SO_4^{2-} and NO_3^{-} between PNA+ and PNA– months were larger in 322 323 the eastern U.S. than in the western U.S.. As shown in Table 1, the differences in the averaged concentrations of SO_4^{2-} , NO_3^{-} and NH_4^{+} were, respectively, 0.1 µg m⁻³ 324 (3.7%), 0.4 µg m⁻³ (36.5%), and 0.1 µg m⁻³ (10.5%) in the eastern U.S., 0.03 µg m⁻³ 325

(3.2%), 0.2 µg m⁻³ (23.8%), and 0.2 µg m⁻³ (31.6%) in the western U.S., as well as 326 0.01 μ g m⁻³ (0.5%), 0.3 μ g m⁻³ (29.1%), and 0.1 μ g m⁻³ (11.9%) in the whole of U.S.. 327 With regard to carbonaceous aerosols, among the 105 and 104 sites with 328 329 measurements of OC and BC, 39% and 31% of which pass the two-tail t-test with 90% 330 confidence, respectively. The differences in concentrations of these two species 331 between PNA+ and PNA- months show similar geographical pattern, with positive 332 values at most sites but negative values in Michigan, New York, and the South 333 Atlantic States. The maximum differences between the PNA+ and PNA- months reached 2.5–3 μ g m⁻³ (50–70%) in Kentucky for OC. Averaged over sites with 334 335 measurements available, the absolute differences in OC and BC concentrations 336 between the PNA+ and PNA- months were larger in the western U.S. than in the 337 eastern U.S. (Table 1). Among all aerosol species listed in Table 1, OC exhibited the 338 largest absolute differences between the PNA phases in the western U.S., because 339 OC accounts for 25–65% of PM_{2.5} in the western U.S. (Malm et al. 2004) and the OC 340 observed by EPA-AQS network, which are located in urban and suburban settings, 341 were higher than the observations by other long term networks in U.S..(Malm et al. 342 2011, Rattigan et al. 2011, Hand et al. 2012b, 2014).

Observations from EPA-AQS datasets indicate the large impacts of PNA phase on aerosol concentrations in the U.S.. It should be noted that, in our analyses above, the locations of measurements and the numbers of samples were different for different aerosol species. The regional averages were also influenced by the uneven distributions of observational sites in different regions. Therefore, model results from the GEOS-Chem simulation will be used to further analyze the impacts of PNA on aerosols in the U.S., as presented in the subsequent sections.

350 We have also calculated the correlation coefficient between PNAI and EPA-AQS 351 surface aerosol concentrations at each site for each aerosol species (PM_{2.5}, SO²⁻₄, NO₃⁻, NH₄⁺, OC, or BC) (see auxiliary Fig. S2). About 40%, 30%, 47%, 33%, 33% and 352 353 34% of sites pass the two-tail t-test with 90% confidence for PM_{2.5}, SO₄²⁻, NO₃⁻, NH₄⁺, 354 OC and BC, respectively. At most sites, positive (negative) correlation coefficients in 355 Fig. S2 corresponded to the increases (decreases) in aerosol concentration in PNA+ 356 months relative to PNA- months shown in Fig. 3. Positive correlation coefficients 357 were large over California, the contiguous Salt Lake, and over and near the eastern Midwest. The fraction of temporal variability explained by PNA (FTVEP) can be 358 359 quantified approximately by the square of correlation coefficient 360 (http://mathbits.com/MathBits/TISection/Statistics2/correlation.htm) (see auxiliary Fig. S3). For all aerosol species, FTVEP were about 5–15% at most sites. For PM_{2.5}, SO²⁻, 361 362 NO_3^- and NH_4^+ aerosols, FTVEP values were high over and near the eastern Midwest, where the PNA teleconnection explained up to 50%, 40%, 50%, and 40% of temporal 363 364 variances of surface concentrations of these aerosol species, respectively.

365

366 4 Impacts of PNA on simulated aerosol concentrations

367 4.1 Simulated aerosol concentrations and model evaluation

Fig. 4a shows the simulated surface-layer concentrations of $PM_{2.5}$ (the sum of SO_4^{2-} , NO₃⁻, NH₄⁺, BC, and OC) and each aerosol species averaged over NDJFM of 1999– 2006. These years are selected because they are the common years of model results and EPA-AQS observation datasets. The simulated PM_{2.5} concentrations were higher in the eastern U.S. than in the western U.S.. The maximum surface PM_{2.5} concentrations reached 14–16 µg m⁻³ in Ohio and Pennsylvania. PM_{2.5} concentrations in the western U.S. were generally less than 4 µg m⁻³, except for

California where $PM_{2.5}$ concentrations were 2–6 µg m⁻³. The distribution of SO_4^{2-} was 375 similar to that of PM_{2.5}, with higher concentrations in the eastern U.S. $(1-8 \ \mu g \ m^{-3})$ 376 than in the western U.S. $(0-3 \ \mu g \ m^{-3})$ due to the coal-fired power plants in the 377 378 Midwest (Park et al. 2006). The concentrations of NO_3^- and NH_4^+ were the highest over and near the eastern Midwest, with values of 3-4 and $2-3 \ \mu g \ m^{-3}$, respectively. 379 The maximum OC concentrations were simulated to be 2–3 μ g m⁻³ in two regions, 380 381 from Ohio to Massachusetts and from Alabama to South Carolina. The simulated BC concentrations in the U.S. were $0-1 \ \mu g \ m^{-3}$, except for the contiguous New York 382 where BC concentrations reached 1–2 μ g m⁻³. The magnitudes and geographic 383 distributions of SO_4^{2-} , NO_3^{-} , NH_4^{+} concentrations simulated in our work are similar to 384 those simulated by Park et al. (2006) and Pye et al. (2009), and our simulated OC 385 386 and BC were similar to those reported by Park et al. (2003).

387 Figure 4b presents the scatter plots of the simulated concentrations versus the EPA-AQS observations. The simulated PM_{2.5} concentrations had normalized mean 388 bias $(NMB = \sum_{m=1}^{M} (S_m - O_m) / \sum_{m=1}^{M} (O_m) \times 100\%$, where S_m and O_m are the 389 390 simulated and observed aerosol concentrations in month m, respectively. M is the 391 total number of winter months examined) of -30% over the U.S., and the correlation coefficient between simulated and observed $\mathsf{PM}_{2.5}$ concentrations was 0.57. The 392 simulated wintertime SO_4^{2-} , NO_3^- and NH_4^+ had NMBs of 37%, 4%, and 26%, 393 respectively. Similar bias in simulated SO_4^{2-} in December-January-February (DJF) 394 395 was reported Park et al. (2006), as the GEOS-Chem model results were compared 396 with observations from the Clean Air Status and Trends Network (CASTNET). The high bias in our simulated NH_4^+ was associated with the overestimation of SO_4^{2-} . Our 397 model underestimates $PM_{2.5}$, SO_4^{2-} , NO_3^{-} , NH_4^{+} , OC and BC in the western U.S. (Fig. 398

399 4b), which can be explained in part by the relatively high aerosol concentrations observed for this region from the EPA-AQS. Hand et al. (2014) compared the 400 401 observed concentrations of aerosols from the EPA-AQS with those from the 402 Interagency Monitoring of Protected Visual Environments (IMPROVE) for 2008–2011, 403 and showed that the ratios of wintertime aerosol concentrations of ammonium sulfate, 404 ammonium nitrate, OC, and BC from the EPA-AQS to those from the IMPROVE were, 405 respectively, 2.3, 7.7, 8.3, and 13.1, as the concentrations were averaged over the 406 western U.S.. Liu et al. (2004) also attributed the high EPA-AQS concentrations in the 407 western U.S. to the relative sparse urban sites that were heavily influenced by strong 408 local sources such as automobiles and wood fires. The low model biases in the 409 western U.S. may also be caused by the biases in emissions in the model.

410 Since this study is dedicated to examine the influence of PNA phase on the 411 month-to-month variations of aerosol concentrations during wintertime, Fig. 4c 412 compares, for each aerosol species, the deviation from the mean (DM) of observed 413 concentration with that of simulated concentration for each winter month. The DM is defined as $DM_m = \left(C_m - \frac{1}{M}\sum_{m=1}^M C_m\right) / \frac{1}{M}\sum_{m=1}^M C_m$, where C_m is the simulated 414 415 average aerosol concentration over the U.S. in month m, and M is the number of 416 winter months examined (we consider the months of NDJFM over 1999-2006 for $PM_{2.5}$, and the months of NDJFM over 2000–2006 for SO_4^{2-} , NO_3^{-} , NH_4^{+} , BC, and OC). 417 The model captures fairly well the peaks and troughs of DMs for PM_{2.5}, SO₄²⁻, NO₃⁻ 418 419 and NH⁺₄, with correlation coefficients of 0.61, 0.45, 0.33, and 0.65, respectively. The 420 model does not capture well the monthly variations of DMs of concentrations of OC 421 and BC, because both anthropogenic and biomass burning emissions are fixed at 422 year 2005 levels during our simulation over 1986-2006 to isolate the impacts of variations in meteorological parameters (PNA phases) on aerosols (see Sect. 2.2). 423

424 Since the biomass burning emissions, which contribute largely to carbonaceous 425 aerosols, have large interannual variations (Duncan et al., 2003; Generoso et al., 426 2003; van der Werf et al., 2006), we also show in Fig. 4c the time series of DMs of 427 biomass burning emissions of OC and BC by using biomass burning emissions in 428 NDJFM over 2000-2006 from GFED v2. The correlation coefficients between 429 biomass burning emissions and observed concentrations of OC and BC were 0.36 430 and 0.34, respectively, indicating that the observed variations in OC and BC were 431 influenced by monthly and interannual variations in biomass burning. We have also 432 calculated the temporal correlation coefficient between EPA-AQS observations and 433 GEOS-Chem model results at each site for each aerosol species (auxiliary Fig. S4). The temporal correlations were statistically significant for PM_{2.5}, SO₄²⁻, NO₃⁻, NH₄⁺ at 434 435 most sites in the U.S., especially over and near the eastern Midwest where largest 436 increases in aerosol concentrations were identified in the PNA+ months relative to the 437 PNA- months.

438

439 **4.2** Impact of PNA on simulated surface-layer aerosol concentrations

We have performed the GEOS-Chem simulation for years 1986–2006, in which there were 35 PNA+ and 35 PNA– months (Fig. 2). Figure 5a shows the concentrations of PM_{2.5} and each aerosols species (SO_4^{2-} , NO_3^{-} , NH_4^{+} , BC, and OC) averaged over the PNA– months of 1986–2006. The magnitudes and geographic distributions of aerosol concentrations in PNA– months were similar to those averaged over NDJFM of years 1999–2006 in Fig. 4.

The simulated absolute and relative differences in aerosol concentrations between PNA+ and PNA– months are shown in Figs. 5b and 5c. The $PM_{2.5}$ concentrations over the U.S. are simulated to increase in PNA+ relative to PNA–

449 months. The maximum enhancement in PM_{2.5} concentrations in PNA+ months was 1.8–2.4 µg m⁻³ (20–40%), located in the juncture of Tennessee and Arkansas. Note 450 that the pattern of simulated differences in PM2.5 between PNA+ and PNA-months 451 452 was similar to that of observations (Fig. 3), except that the simulated differences were 453 not large in California, mainly due to the underestimation of OC in California as 454 compared to EPA-AQS data (Fig. 4b). The simulated PM_{2.5} concentrations were higher by 0.6 μ g m⁻³ (12.2%), 0.3 μ g m⁻³ (14.0%), and 0.9 μ g m⁻³ (10.8%) over the 455 456 whole of, western, and eastern U.S., respectively, in the PNA+ months than in PNAmonths (Table 2). The simulated relative difference in PM_{2.5} was close to that from 457 458 observations (Table 1) in the western U.S., but the simulated relative differences in PM_{2.5} were larger than those from observations in the eastern and whole of U.S.. 459

460 Fig. 5 also shows the differences in simulated surface-layer concentrations of SO_4^{2-} , NO_3^{-} and NH_4^{+} between the PNA+ and the PNA– months. The differences in 461 concentrations of SO_4^{2-} were larger in the western than in the eastern U.S., with 462 maximum enhancements of 0.4–0.8 μ g m⁻³ (30–50%) over the West North Central 463 464 States (South Dakota, Nebraska, Minnesota, Iowa, and Missouri). The differences in 465 concentrations of NO_3^- and NH_4^+ between PNA+ and PNA– months had similar 466 geographical patterns, with increases in concentrations in a large fraction of the 467 eastern U.S. and over a belt region along the Rocky Mountain in the western U.S.. The increases in concentrations of NO₃⁻ and NH₄⁺ over the eastern U.S. in PNA+ 468 469 months relative to PNA- months agreed very well with those seen in observations in 470 most regions of the U.S. (Fig. 3). The model underestimates the differences in NO_3^- 471 in California and the southeastern U.S., because the model does not capture well the 472 temporal variations of NO₃⁻ in these two regions (Fig. S4). The differences in

473 concentrations of SO_4^{2-} , NO_3^{-} and NH_4^{+} in the eastern U.S. between the PNA+ and 474 PNA– months were 0.2 µg m⁻³ (4.0%), 0.4 µg m⁻³ (33.5%), and 0.2 µg m⁻³ (13.2%), 475 respectively (Table 2).

476 The differences in concentrations of OC and BC between PNA+ and PNA-477 months had similar geographical patterns, with large increases in concentrations over 478 and near the eastern Midwest and the region from northwestern U.S. to Texas. The maximum differences reached 0.2–0.4 μ g m⁻³ (10–20%) and 0.1–0.2 μ g m⁻³ (20– 479 480 30%) in Illinois, Indiana and Ohio for OC and BC, respectively. The magnitudes of the 481 differences in OC and BC were statistically significant but were smaller than the 482 observations (Tables 1 and 2). The absolute differences in OC were less than 0.1 µg m⁻³ in the western, eastern and whole of U.S due to the underestimation of OC in the 483 484 simulation.

485 In summary, model results agreed with observations in that the concentrations of all aerosol species of SO₄²⁻, NO₃⁻, NH₄⁺, BC, and OC averaged over the U.S. were 486 487 higher in PNA+ months than in PNA- months. Relative to the PNA- months, the 488 average concentration of PM_{2.5} over the U.S. was higher by about 8.7% (12.2%) 489 based on observed (simulated) concentrations. Furthermore, simulated geographical 490 patterns of the differences in PM_{2.5} and each aerosol species between the PNA+ and 491 PNA- months were similar to those seen in observations in most areas of the U.S. 492 (except for California), with the largest increases in aerosol concentrations in PNA+ 493 months over and near the eastern Midwest.

494

495 **5** Mechanisms for the impact of PNA on aerosol concentrations

496 **5.1** The impact of PNA on transboundary transport of aerosols

497 The transboundary transport of pollutants to and from the U.S. depends largely on 498 winds in the free troposphere (Liang et al. 2004). Figure 6 shows the horizontal winds 499 at 700 hPa averaged over the winter months of NDJFM of 1986-2006 and the 500 corresponding differences between PNA+ and PNA- months on the basis of the 501 GEOS-4 meteorological fields. Strong westerlies prevailed over the U.S. in wintertime 502 (Fig. 6a). Relative to the PNA- months, anomalous northeasterlies occurred over a large fraction of U.S. in the PNA+ months. Anomalous anti-cyclonic circulation 503 504 occurred near the northwestern U.S. and anomalous cyclonic circulation occurred 505 near the southeastern U.S. (Fig. 6b), corresponding to the large positive and negative 506 differences in geopotential height in these two regions (see auxiliary Fig. S1), 507 respectively.

508 We also calculate mass fluxes of PM_{2.5} at the four lateral boundaries (from the 509 surface to 250 hPa) of the U.S. for different PNA phases (Table 3). The domain of the 510 box to represent the U. S. is (75–120°W, 28–49°N), as shown in Fig. 6b. For PM_{2.5} in 511 wintertime, the inflow from the west boundary and the outflow from the east boundary 512 had the largest absolute values (Table 3). Relative to the PNA- months, the inflow 513 from the west boundary and the outflow from the east boundary in PNA+ months exhibited reductions of 16.1 kg s⁻¹ and 13.5 kg s⁻¹, respectively (Table 3). The inflow 514 flux from south boundary decreased by 15.8 kg s⁻¹, and the inflow flux from north 515 boundary increased by 31.7 kg s⁻¹, leading to a net increase of inflow flux of 13.3 kg 516 517 s^{-1} in PNA+ months. Therefore, the transboundary transport has an overall effect of 518 increasing PM_{2.5} aerosols in the U.S. in PNA+ months relative to PNA– months. The relative change in net flux was 9.9% ((PNA+ minus PNA-).100%/PNA-) (Table 3), 519

520 coinciding well with the enhancement of 12.2% in surface-layer $PM_{2.5}$ concentration 521 averaged over the U.S. (Table 2). Note that because the GEOS-Chem model 522 underestimates $PM_{2.5}$ concentrations in the western U.S. (Fig. 4b), the net outflow 523 flux from the selected box might have been underestimated, but this should not 524 compromise our conclusions about the relative differences in net flux between PNA+ 525 and PNA– phases.

526 **5.2** Local changes in aerosol concentrations caused by the PNA

527 The PNA pattern is also associated with variations in meteorological variables such 528 as temperature (Leathers et al. 1991; Konrad II 1998; Notaro et al. 2006; Knight et al. 529 2008; Liu et al. 2015; Ning and Bradley 2014, 2015), precipitation (Leathers et al. 530 1991; Henderson and Robinson 1994; Coleman and Rogers 2003; Notaro et al. 2006; 531 Archambault et al. 2008; Myoung and Deng 2009; Ning and Bradley 2014, 2015; 532 Wise et al. 2015) and humidity (Sheridan 2003; Coleman and Rogers 2003; Knight et 533 al. 2008) in U.S., which are expected to influence aerosol concentrations within the 534 U.S. through chemical reactions, transport, and deposition.

Figure 7 shows the composite differences, between the PNA+ and PNA- months, 535 536 in surface air temperature (T), precipitation rate (PR), relative humidity (RH), surface 537 wind speed (WS), and planetary boundary layer height (PBLH), based on the 538 reanalyzed GEOS-4 datasets. Relative to PNA- months, temperatures were higher 539 by 1–3 K over the northwestern U.S. and lower by 1–4 K in the southeastern region. 540 Such geographic distributions of temperature anomalies were attributed to the 541 maritime warm air in the northwestern U.S. accompanied by the enhanced 542 tropospheric geopotential height in North America (Leathers et al. 1991; Sheridan 543 2003; Liu et al. 2015; Ning and Bradley 2015) (see also auxiliary Fig. 1d) and the 544 more frequent outbreak of cold air in southeastern U.S. accompanied by the

545 depressed geopotential height (Konrad II 1998; Liu et al. 2015) (see also auxiliary Fig. 546 1d). The differences in precipitation between PNA+ and PNA- months reached -1.6 to -2.4 mm day^{-1} (-32 to -48%) over and near the eastern Midwest, -2.4 to -3.2 mm547 day^{-1} (-48 to -64%) in the northwestern U.S., and 1.6-2.4 mm day^{-1} (16-32%) in the 548 549 southeastern U.S.. These effects of PNA on precipitation were similar to those obtained from wintertime station data by Leathers et al. (1991), Coleman and Rogers 550 551 (2003) and Wise et al. (2015). With respect to RH, the values in the eastern U.S. were 552 generally lower in the PNA+ months than in PNA- months, as a result of the reduced 553 moisture flux from the Gulf of Mexico to the eastern U.S. (Coleman and Rogers 2003), 554 where RH showed maximum reduction of -3 to -9%. The enhancement of RH of up 555 to 6–9% in Texas, Oklahoma and New Mexico was due to the anomalous easterlies 556 over the south central U.S. (see Fig. 7), which diminished the influence of the dry air 557 from the deserts of the southwestern U.S. and northwestern Mexico in PNA+ months 558 (Sheridan 2003). The surface WS showed reductions in PNA+ months relative to 559 PNA– months in two regions, one was the belt region along the Rocky Mountain from the northwestern U.S. to Texas (with the maximum reductions of 1.5–2.0 m $\rm s^{-1}$ (48– 560 561 64%)) and the other region, with a northeast-southwest orientation, was from Ohio to Louisiana (with maximum reductions of $0.5-1.0 \text{ m s}^{-1}$ (16–32%)). The differences in 562 PBLH between PNA+ and PNA- months were statistically significant in the western 563 564 U.S. and in the belt region from the northeastern U.S. to the eastern Midwest, with the maximum reductions in PBLH of 75-100 m (15-20%) and 75-100 m (10-15%), 565 566 respectively. The changes of PBLH were possibly associated with the frequent 567 outbreak of cold air in the U.S. in PNA+ phases (Leathers et al., 1991; Archambault et al., 2010), which induced cool air at the surface and clouds over the intermountain 568 569 and the eastern Midwest regions (Sheridan, 2003).

570 The comparisons of Fig. 5b with Fig. 7a indicate that the increases in PM_{2.5} 571 concentrations over and near the eastern Midwest in PNA+ months relative to the 572 PNA- months can be attributed to the decreases in PR, WS and PBLH in these 573 locations, since the changes in these three variables depressed wet deposition, local 574 horizontal diffusion, and vertical diffusion of the surface aerosols, respectively. The increases in SO₄²⁻ in the western U.S. (Fig. 5b) corresponded to the decreases in PR 575 576 and PBLH and the increased temperature. The increases in temperature enhance 577 chemical reaction rates (Aw and Kleeman 2003; Dawson et al. 2007; Kleeman 2008). In the eastern U.S., although PR, WS, and PBLH decreased over and near the 578 579 eastern Midwest, the cooling in the eastern U.S. might have offset the effects by PR, WS, and PBLH, inducing practically no changes in SO_4^{2-} in the eastern U.S.. The 580 large increases in NO_3^- and NH_4^+ in the southeastern U.S. (Fig. 5b) can be attributed 581 582 to the reduced surface temperature, which was favorable to the wintertime formation of NO_3^- and NH_4^+ (Dawson et al. 2007). The differences in concentrations of OC and 583 584 BC between PNA+ and PNA- corresponded well with the reduced PR, WS and PBLH. 585

586 In order to quantify the impacts of anomalies in meteorological parameters driven 587 by PNA on concentrations of different aerosol species, the pattern correlation 588 coefficients (PCC. http://glossary.ametsoc.org/wiki/Pattern correlation) are 589 calculated and shown in Table 4. These pattern correlation coefficients denote the 590 relationship between the geographical distribution of anomalies of each of T, PR, RH, 591 WS, and PBLH (Fig. 7b) and that of the differences in concentration of each aerosol 592 species between PNA+ and PNA- months (Fig. 5c). As shown in Table 4, over the whole U.S., the PNA influenced SO_4^{2-} concentrations mainly through changes PBLH, 593 594 PR, and T, with the highest PCC values of -0.43, -0.38, and +0.26, respectively. For

595 NO_{3}^{-} , the PNA-induced variations in temperature had a strong negative correlation 596 (PCC=-0.59) with the PNA-induced differences in concentrations, indicating that 597 surface temperature was the dominant meteorological factor to influence NO_{3}^{-} 598 concentrations. For NH_{4}^{+} , OC, and BC, PR and PBLH were the two variables that had 599 the largest negative PCC values (Table 4).

600

601 6 Conclusions

This study examined the impacts of monthly variation in the PNA phase on wintertime surface-layer aerosol concentrations in the U.S. by the analyses of EPA-AQS observations over 1999–2013 and model results for 1986–2006 from the global chemical transport model GEOS-Chem.

606 The composite analyses on the EPA-AQS observations showed that the average concentrations of $PM_{2.5}$, SO_4^{2-} , NO_3^{-} , NH_4^{+} , OC, and BC aerosols over the U.S. were 607 higher in the PNA+ months than in the PNA- months by 1.0 μ g m⁻³ (8.7%), 0.01 μ g 608 m^{-3} (0.5%), 0.3 µg m^{-3} (29.1%), 0.1 µg m^{-3} (11.9%), 0.6 µg m^{-3} (13.5%), and 0.2 µg 609 m^{-3} (27.8%), respectively. Regionally, the observed $PM_{2.5}$ concentrations were higher 610 by 3–5 μ g m⁻³ (40–80%) over the Midwest, and by 7–9 μ g m⁻³ (80–100%) around the 611 612 Salt Lake, as the concentrations in PNA+ months were compared to those in PNA-613 months.

The impacts of PNA phase on aerosol concentrations were reproduced fairly well by the GEOS-Chem simulation with fixed anthropogenic emissions (the variations of aerosols concentrations were driven by changes in meteorological fields alone). Concentrations of SO_4^{2-} , NO_3^{-} , NH_4^{+} , BC, and OC averaged over the U.S. were simulated to be higher in the PNA+ months than in PNA– months. The average concentration of $PM_{2.5}$ over the U.S. was simulated to be 12.2% higher in the PNA+

620 months relative to the PNA– months, in close agreement with the observations. 621 Simulated geographical patterns of the differences in PM_{2.5} and each aerosol species 622 between the PNA+ and PNA– months were similar to those seen in observations. The 623 largest increases in aerosol concentrations in the PNA+ months were simulated to be 624 over and near the eastern Midwest, but the model results showed small PNA-induced 625 changes in aerosol concentrations in the western U.S..

626 The mechanisms for the impacts of PNA on aerosol concentrations were 627 examined. The transboundary transport was found to have an overall effect of increasing PM_{2.5} aerosols in the U.S. in the PNA+ months relative to PNA– months. 628 629 Compared to the PNA- months, anomalous northeasterlies occurred over a large 630 fraction of U.S., which led to a net increase in inflow flux of $PM_{2.5}$ of 14.0 Kg s⁻¹ in 631 PNA+ months. Regionally within the U.S., the PNA influenced aerosol concentrations 632 through changes in precipitation rate (PR), planetary boundary layer height (PBLH), 633 surface wind speed (WS), surface air temperature (T) and relative humidity (RH), as 634 represented by the pattern correlation coefficients (PCCs). The PNA influenced SO₄²⁻ 635 concentration mainly through changes PBLH, PR, and T, with the highest PCC values 636 of -0.43, -0.38, and +0.26, respectively. For NO₃, the PNA-induced variations in 637 temperature had a strong negative correlation (PCC=-0.59) with the PNA-induced 638 differences in concentrations. For NH⁺₄, OC, and BC, PR and PBLH were the two 639 variables that had the largest negative PCC values. It should be noted that, the PCC 640 values only statistically present the relationship between meteorological parameters 641 and aerosol concentrations in the U.S.. More in-depth understanding of the impact of 642 PNA phase on aerosol concentrations should be carried out on the basis of physical 643 and chemical processes.

644 Conclusions from this study have important implications for air quality in the U.S.. 645 Leathers and Palechi (1992) showed that the PNAI were generally low in 1947–1957 646 but consistently high in 1958–1987. The PNAI during 1948–2010 exhibited an 647 increasing trend for positive phases and a decreasing trend for negative phases (Liu 648 et al. 2015; Ning et al. 2015; http://research.jisao.washington.edu/data sets/pna/#dif). 649 indicating that wintertime particulate matter pollution in most areas of U.S. 650 deteriorated due to variations in PNA phase alone. Climate models projected that 651 positive PNA phases would increase in the future because of the global warming 652 (Kachi and Nitta, 1997; Müller and Roeckner 2008; Zhou 2014). Therefore, the trend 653 in PNA pattern underlies the necessity of strict emission reduction strategies for 654 greenhouse gases, aerosols, and aerosol precursors.

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Table 1. The absolute (μ g m⁻³) and relative (%) differences in observed aerosol concentrations between the PNA+ and PNA– months (PNA+ minus PNA–). The observed concentrations are averaged over all the sites in the whole of U.S., in the western U.S., or in the eastern U.S.. See Fig 1 for locations of the sites. The measurements are from the EPA-AQS data. The ** and * indicate the differences that have passed the two-tail student-t test with 95% and 90% significance levels, respectively.

- ,			
	Whole U.S.	Western U.S.	Eastern U.S
PM _{2.5}	1.0 (8.7%)**	1.3 (14.3%)**	0.8 (7.2%)**
SO_4^{2-}	0.01 (0.5%)	0.03 (3.2%)	0.1 (3.7%)
NO_3^-	0.3 (29.1%)**	0.2 (23.8%)**	0.4 (36.5%)**
NH_4^+	0.1 (11.9%)*	0.2 (31.6%)**	0.1 (10.5%)
OC	0.6 (13.5%)**	0.9 (17.7%)**	0.3 (8.0%)**
BC	0.2 (27.8%)**	0.2 (25.0%)**	0.1 (25.2%)**

Table 2. The absolute (μ g m⁻³) and relative (%) differences in simulated aerosol concentrations between the PNA+ and PNA– months (PNA+ minus PNA–). The simulated concentrations are averaged over the whole of U.S., the western U.S. (west of 100°W), or the eastern U.S. (east of 100°W). The concentrations are from the GEOS-Chem simulation for 1986–2006. The ** and * indicate the differences that have passed the two-tail student-t test with 95% and 90% significance levels, respectively.

	Whole U.S.	Western U.S.	Eastern U.S.
PM _{2.5}	0.6 (12.2%)**	0.3 (14.0%)**	0.9 (10.8%)**
SO_4^{2-}	0.2 (7.1%)	0.1 (13.5%) **	0.2 (4.0%)
NO_3^-	0.2 (30.3%) **	0.1 (28.5%) **	0.4 (33.5%)**
NH_4^+	0.2 (14.4%)**	0.1 (15.4%)**	0.2 (13.2%)**
OC	0.05 (6.5%)**	0.03 (8.6%)**	0.08 (5.9%)**
BC	0.03 (10.2%)**	0.01 (8.6%)**	0.05 (11.0%)**

Table 3. The composite analyses of horizontal mass fluxes (kg s⁻¹) of PM_{2.5} for the selected box of (75–120°W, 28–49°N, from the surface to 250 hPa) in the PNA+ and PNA– months of 1986–2006. The positive values at the four boundaries indicate eastward or northward transport, and negative values indicate westward or southward transport. The positive (negative) value of net flux indicates the net gain (loss) of PM_{2.5} in the selected box. All the fluxes are from the GEOS-Chem simulation.

	Boundaries and total	Mass Flux
	West	75.0
	East	233.9
PNA+	South	15.0
	North	-22.9
	Net flux	-120.9
	West	91.1
	East	247.4
PNA-	South	30.8
	North	8.8
	Net flux	-134.2
	West	-16.1
Diff	East	-13.5
DIII. (DNA + minuo DNA -)	South	-15.8
(FINAT IIIIIIUS FINA-)	North	-31.7
	Net flux	13.3

Table 4. The pattern correlation coefficients between the composite differences in aerosol concentrations (Fig. 5c) and the corresponding composite differences in meteorological parameters (Fig. 7b). The * denotes the correlations that have passed the two-tail t-test with 95% confidence level.

	PM _{2.5}	SO ₄ ²⁻	NO_3^-	NH_4^+	OC	BC
Т	-0.13	0.26*	-0.59*	-0.22*	0.07	-0.16
PR	-0.44*	-0.38*	0.04	-0.42*	-0.63*	-0.50*
RH	-0.08	-0.05	-0.02	0.12	-0.32*	-0.36*
WS	-0.27*	-0.1	-0.22*	-0.27*	-0.28*	-0.24*
PBLH	-0.61*	-0.43*	-0.32*	-0.61*	-0.60*	-0.55*

1093 **Figure Captions** 1094

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- **Fig. 1.** The locations of EPA-AQS sites with measurements that meet the criteria described in Sect. 2.1 in the text. $PM_{2.5}$ measurements are available for years of 1999–2013 (sites are marked by black dots) and speciated aerosol ($SO_4^{2^-}$, NO_3^- , NH_4^+ , BC, and OC) concentrations are available over years of 2000–2013 (sites are marked by red diamonds). The grey solid line defines the western United States (west of 100°W) and eastern United States (east of 100°W).
- 1103 Fig. 2. (a) PNAI for years of 1986-2013 calculated using the NCEP-2 data (NCEP2-PNAI), with the PNA+ and PNA- months during 1999-2013 indicated. 1104 (b) PNAI for years of 1986–2013 calculated using the NCEP-2 data, with the 1105 PNA+ and PNA- months during 2000-2013 indicated. (c) PNAI for years of 1106 1107 1986-2006 calculated using the GEOS-4 data (GEOS4-PNAI), with the PNA+ and PNA- months over 1986-2006 indicated. Red circles are PNA+ months 1108 and blue circles PNA- months. The PNA+ (PNA-) months are defined as the 1109 1110 25% of winter months examined, which have the highest positive (negative) 1111 PNA index values. 1112
- **Fig. 3.** The absolute (μ g m⁻³, left column) and relative differences (%, right column) in 1113 1114 observed monthly mean aerosol concentrations between PNA+ and PNA-1115 months (PNA+ minus PNA-). See Fig. 2c for the definitions of PNA+ and 1116 PNA- months. The measurements of PM_{2.5} were carried out over 1999–2013, in which there were 18 PNA+ months and 18 PNA- months as shown in Fig. 1117 1118 2a. The measurements of speciated aerosols were taken during 2000–2013, in 1119 which there were 17 PNA+ and 17 PNA– months (Fig. 2b). The seasonal cycle 1120 and trend in observed aerosol concentrations were removed as described in 1121 Sect. 2.1. The sites with black dots were the differences that passed the 1122 two-tail t-test with 90% confidence level.
- Fig. 4. (a) Simulated surface-layer concentrations ($\mu g m^{-3}$) of PM_{2.5} (the sum of SO₄²⁻, 1124 NO_{3}^{-} , NH_{4}^{+} , BC, and OC) and each aerosol species averaged over NDJFM of 1125 1126 1999–2006. (b) Scatter plots of the simulated concentrations (ug m⁻³, vertical axis) versus the EPA-AQS observations ($\mu g m^{-3}$, horizontal axis). Also shown 1127 are the y=x line (black dash line), linear fit for whole U.S. (black line), linear fit 1128 1129 for western U.S. (blue line), and linear fit for eastern U.S. (red line). The blue 1130 and red dots represent sites in the western and eastern U.S., respectively, (c) 1131 Comparisons of the deviation from the mean (DM) of observed concentration (black line) with that of simulated concentration (red line) in each winter month 1132 1133 for each aerosol species, left axis. Also shown in the panel for OC (BC) the 1134 monthly variation in DM of biomass burning emission of OC (BC), blue line, 1135 right axis. 1136
- Fig. 5. (a) Simulated concentrations (μg m⁻³) of PM_{2.5} and each aerosols species averaged over the PNA– months of 1986–2006. (b) The absolute differences (μg m⁻³) in simulated aerosol concentrations between PNA+ and PNA– months (PNA+ minus PNA–). (c) The relative differences (%) in simulated aerosol concentrations between PNA+ and PNA– months. The white spaces in (C)

indicate the areas that did not pass the two-tail student-t test with 90%
significance level. The seasonal cycles of simulated aerosol concentrations
were removed. See Fig. 2c for the definitions of PNA+ and PNA- months.

- Fig. 6. (a) Horizontal winds at 700 hPa averaged over the winter months of NDJFM of 1986–2006, and (b) the corresponding differences between the PNA+ and PNA- months. Datasets are from the assimilated GEOS-4 meteorological fields. Also shown in (b) is the domain of (75–120°W, 28–49°N) for which transboundary mass fluxes of PM_{2.5} are calculated. See Fig. 2c for the definitions of PNA+ and PNA- months.
- Fig. 7. (a) The absolute and (b) relative differences in meteorological parameters between PNA+ and PNA- months. Datasets are from the assimilated GEOS-4 meteorological fields. The white spaces in indicate the areas that did not pass the two-tail student-t test with 90% significance level. The seasonal cycles of meteorological variables were removed, similar to the treatment for observations in Fig. 3. See Fig. 2c for the definitions of PNA+ and PNA- months.



Fig. 1. The locations of EPA-AQS sites with measurements that meet the criteria described in Sect. 2.1 in the text. $PM_{2.5}$ measurements are available for years of 1999–2013 (sites are marked by black dots) and speciated aerosol (SO_4^{2-} , NO_3^{-} , NH_4^+ , BC, and OC) concentrations are available over years of 2000–2013 (sites are marked by red diamonds). The grey solid line defines the western United States (west of 100°W) and eastern United States (east of 100°W).



1168 1169

1170 Fig. 2. (a) PNAI for years of 1986-2013 calculated using the NCEP-2 data (NCEP2-PNAI), with the PNA+ and PNA- months during 1999–2013 indicated. (b) 1171 1172 PNAI for years of 1986–2013 calculated using the NCEP-2 data, with the PNA+ and PNA- months during 2000-2013 indicated. (c) PNAI for years of 1986-2006 1173 calculated using the GEOS-4 data (GEOS4-PNAI), with the PNA+ and PNA- months 1174 1175 over 1986-2006 indicated. Red circles are PNA+ months and blue circles PNA-1176 months. The PNA+ (PNA-) months are defined as the 25% of winter months 1177 examined, which have the highest positive (negative) PNA index values.



Fig. 3. The absolute (μ g m⁻³, left column) and relative differences (%, right column) in 1179 observed monthly mean aerosol concentrations between PNA+ and PNA- months 1180 (PNA+ minus PNA-). See Fig. 2c for the definitions of PNA+ and PNA- months. The 1181 measurements of PM_{2.5} were carried out over 1999-2013, in which there were 18 1182 PNA+ months and 18 PNA- months as shown in Fig. 2a. The measurements of 1183 speciated aerosols were taken during 2000-2013, in which there were 17 PNA+ and 1184 1185 17 PNA- months (Fig. 2b). The seasonal cycle and trend in observed aerosol 1186 concentrations were removed as described in Sect. 2.1. The sites with black dots 1187 were the differences that passed the two-tail t-test with 90% confidence level.





Fig. 4. (a) Simulated surface-layer concentrations ($\mu g m^{-3}$) of PM_{2.5} (the sum of SO₄²⁻, 1189 NO₃, NH₄, BC, and OC) and each aerosol species averaged over NDJFM of 1999-1190 2006. (b) Scatter plots of the simulated concentrations ($\mu g m^{-3}$, vertical axis) versus 1191 the EPA-AQS observations ($\mu g m^{-3}$, horizontal axis). Also shown are the y=x line 1192 (black dash line), linear fit for whole U.S. (black line), linear fit for western U.S. (blue 1193 line), and linear fit for eastern U.S. (red line). The blue and red dots represent sites in 1194 the western and eastern U.S., respectively. (c) Comparisons of the deviation from the 1195 1196 mean (DM) of observed concentration (black line) with that of simulated concentration 1197 (red line) in each winter month for each aerosol species, left axis. Also shown in the

panel for OC (BC) the monthly variation in DM of biomass burning emission of OC (BC), blue line, right axis.



1200

Fig. 5. (a) Simulated concentrations ($\mu g m^{-3}$) of PM_{2.5} and each aerosols species 1201 averaged over the PNA– months of 1986–2006. (b) The absolute differences ($\mu g m^{-3}$) 1202 in simulated aerosol concentrations between PNA+ and PNA- months (PNA+ minus 1203 1204 PNA-). (c) The relative differences (%) in simulated aerosol concentrations between 1205 PNA+ and PNA– months. The white spaces in (C) indicate the areas that did not pass 1206 the two-tail student-t test with 90% significance level. The seasonal cycles of simulated aerosol concentrations were removed, similar to the treatment for 1207 1208 observations in Fig. 3. See Fig. 2c for the definitions of PNA+ and PNA- months.



Fig. 6. (a) Horizontal winds at 700 hPa averaged over the winter months of NDJFM of 1986-2006, and (b) the corresponding differences between the PNA+ and PNA-1211 months. Datasets are from the assimilated GEOS-4 meteorological fields. Also shown 1212

- 1213 in (b) is the domain of (75–120°W, 28–49°N) for which transboundary mass fluxes of
- PM_{2.5} are calculated. See Fig. 2c for the definitions of PNA+ and PNA- months. 1214



Fig. 7. (a) The absolute and (b) relative differences in meteorological parameters between PNA+ and PNA– months. Datasets are from the assimilated GEOS-4 meteorological fields. The white spaces in indicate the areas that did not pass the two-tail student-t test with 90% significance level. The seasonal cycles of meteorological variables were removed, similar to the treatment for observations in Fig. 3. See Fig. 2c for the definitions of PNA+ and PNA– months.