Manuscript # ACP-2015-804

Responses to Reviewer #1

General comments. The authors have conducted an interesting study to investigate the influence of the Pacific-North America teleconnection (PNA) on U.S. winter aerosol concentrations using both statistical methods and a chemistry model. This work contributes to our understanding of climate patterns responsible for PM2.5 variability and is appropriate for ACP. But some of the conclusions are relatively weak. So I recommend that the paper can be published only after a major revision as describe below.

1. Even though PNA is the leading circulation pattern in the North Pacific and North America in the troposphere during the wintertime, the authors should also show it is important for the aerosol variability. What is the fraction of temporal variability that can be explained by PNA? To show this, the authors can calculate the correlation coefficient between PNA and aerosol concentrations in each site.

Response:

Following the Reviewer's suggestion, we have calculated the correlation coefficient between PNAI and EPA-AQS surface aerosol concentrations at each site for each aerosol species ($PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, OC, or BC) (auxiliary Fig. S2, see below). At most sites, positive (negative) correlation coefficients in Fig. S2 correspond to the increases (decreases) in aerosol concentration in PNA+ months relative to PNA– months shown in Fig. 3 of our manuscript. Large positive correlation coefficients are found over California, the contiguous Salt Lake, and over and near the eastern Midwest (Fig. S2). The fraction of temporal variability explained by PNA (FTVEP) can be quantified approximately by the square of correlation coefficient (http://mathbits.com/MathBits/TISection/Statistics2/correlation.htm) (auxiliary Fig. S3, see below). For all aerosol species, FTVEP are about 5–15% at most sites. For $PM_{2.5}$, SO_4^{2-}, NO_3^- and NH_4^+ aerosols, high FTVEP values are found over and near the eastern Midwest, where the PNA teleconnection explains up to 50%, 40%, 50%, and 40% of temporal variances of surface concentrations of these aerosol species, respectively. We have added these descriptions to the end of Sect. 3.1.

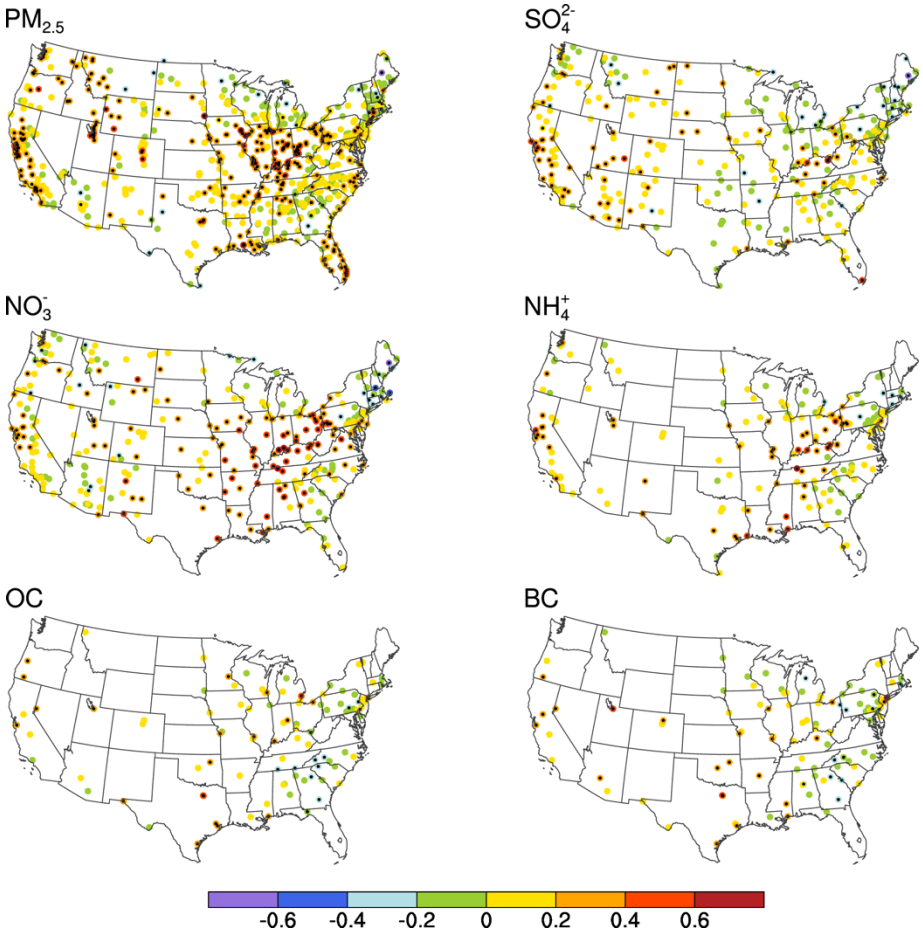

Fig. S2. Correlation coefficient between PNAI and EPA-AQS surface aerosol concentrations at each site for each aerosol species ($PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, BC, or OC). The measurements are the same as those used in Fig. 3. The sites with black dots are those that have passed the two-tail t-test with 90% confidence level.

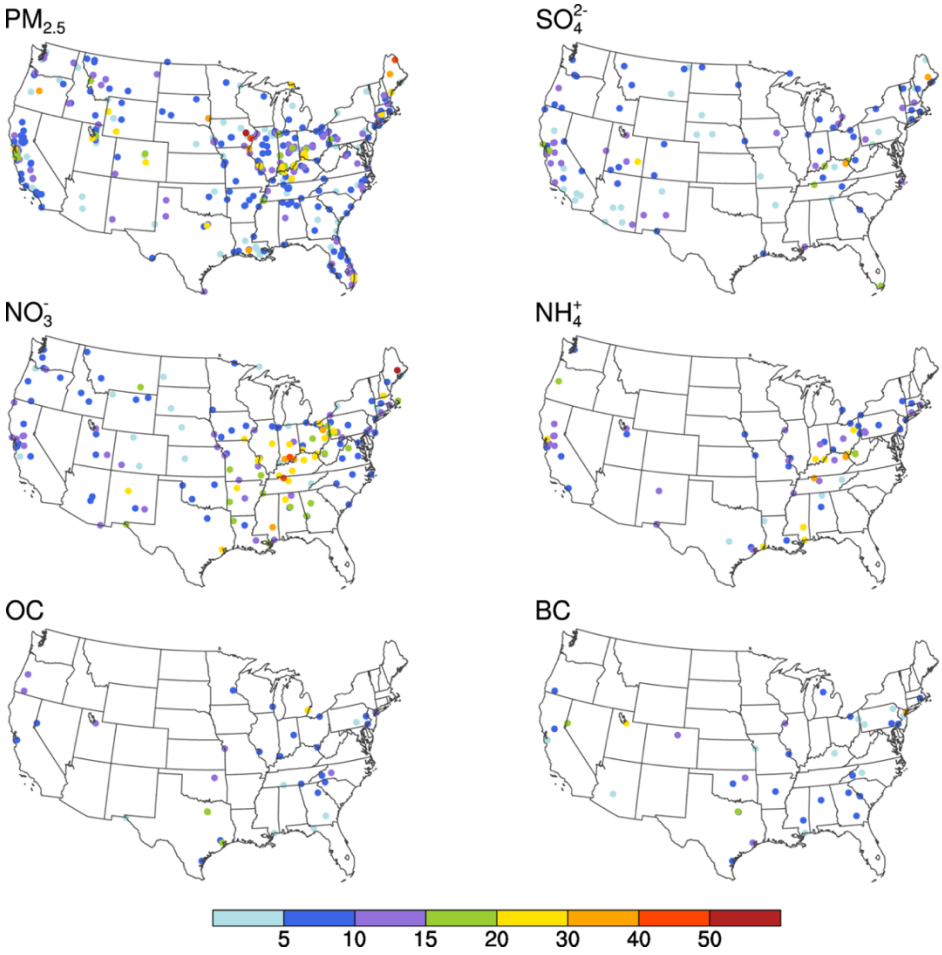

Fig. S3. The fraction (%) of temporal variability of EPA-AQS surface aerosol concentrations explained by PNA at each site for each aerosol species ($PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, BC, or OC). The measurements are the same as those used in Fig. 3. The sites shown here are those in Fig. S2 with the correlation coefficients passed the two-tail t-test with 90% confidence level.

2. Have the authors detrended the PM2.5 observations when they do a composite analysis between positive and negative PNA? The aerosol concentrations have shown a significant decreasing trend from 1999 to present in the United States. Won't this affect the conclusion?

Response:

This is a good point. We have redone the composite analyses for observed aerosol concentrations by detrending the observations from the EPA-AQS. The revised differences in aerosol concentrations between positive and negative PNA are similar to the results in our previous version of manuscript (those obtained without detrending); the horizontal distributions are about the same but the magnitudes of the differences in aerosol concentrations are slightly smaller. We have updated Figs. 3, 4 and Table 1, and have changed the descriptions in the text accordingly.

3. It is hasty, with just some statistical analysis, to conclude that PBL should be the most important meteorological factor that influences the concentrations of PM2.5. Maybe the authors should do a sensitivity test by fixing the PBL height in the chemistry model. If the monthly PM2.5 variability is largely reduced, the major conclusion of this study should be correct.

Response:

We conclude that PBL is the most important meteorological factor that influences the concentrations of $PM_{2.5}$ on the basis of the magnitude of the pattern correlation coefficients (PCC). The same statistical approach was also used in previous studies (Levy et al., 2008; Jeong and Park, 2013; Allen et al., 2015). Levy et al. (2008), by simulations with the NOAA/GFDL climate model, reported that the regional pattern of changes in surface temperature did not correspond well to the regional pattern of radiative forcings of short-lived species because of the low global PCC of –0.172 under the IPCC A1B scenario. Jeong and Park (2013) used the PCC values to show the impacts of variabilities in temperature, precipitation, relative humidity, mixing depth, cloud fraction, and surface wind speed on regional patterns of concentrations of O_3, $(NH_4)_2SO_4$, and NH_4NO_3 in East Asia on the basis of the GEOS-Chem simulation. Allen et al. (2015) quantified the role of precipitation in the changes of aerosol burdens from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP); they obtained global PCC values of –0.36, –0.33 and –0.30 between precipitation and burdens of SO_4^{2-}, BC, and OC, respectively.

The sensitivity test suggested by the Reviewer was carried out by Dawson et al. (2007). Dawson et al. (2007) reported, by fixing all meteorological parameters but perturbing PBLH in the Particulate Matter Comprehensive Air Quality Model with extensions (PMCAMx), that $PM_{2.5}$ concentrations over the Mideast in January decreased by about 1 μg m^{-3} if PBLH decreased by 150 m. They concluded that the effect of mixing height on $PM_{2.5}$ concentrations is rather important, especially in polluted areas. However, such sensitivity experiment by changing one specific meteorological variable (for example, PBLH) ignored the covariance of other meteorological parameters (such as temperature and convection associated with PBLH), which could not give us realistic responses of aerosol concentrations to changes in meteorological conditions.

4. Is there a specific reason that the authors use a very old version of GEOS-Chem model? According to the info here (http://acmg.seas.harvard.edu/geos/), the current public release is v10-01. But the authors still use v8-2 that was released five years ago. Also their model is driven by GEOS4 for 1986-2006. Why not use updated meteorological fields? How does this model treat the Secondary Organic Aerosol (SOA) and what mechanism is used? In the wintertime, SOA might still be a very important component in the south US.

Response:

The GEOS-Chem Version 8-02-01 is used in our study, because we have evaluated this version's performance in simulating aerosol concentrations in

our previous studies (Zhu et al., 2012; Yang et al., 2015). This version has also been used by other recent studies on atmospheric chemistry (Jiang et al. 2015; Luo et al. 2015; Marlier et al. 2015).

Our simulation is driven by the GEOS-4 meteorological data, which are available for years 1986–2006. Although the relatively new MERRA meteorological fields can be used to drive the GEOS-Chem model for years 1979–2013, there is a problem of long-term inhomogeneity in MERRA reanalysis data for precipitation, humidity, and temperature in the lower troposphere due to the introduction of new observation types in different time (http://gmao.gsfc.nasa.gov/pubs/docs/Chen480.pdf), which would influence the decadal simulation in our study. This problem will be solved in MERRA-2 datasets (Molod et al. 2015), but unfortunately the MERRA-2 meteorological fields are currently not available.

Considering that the impacts of PNA on temperature, relative humidity, surface wind speed, and precipitation obtained from the GEOS-4 meteorological data for 1986–2006 (Fig. 7 in manuscript) are similar to those obtained from the NCEP Reanalysis and Global Precipitation Climatology Center (GPCC) data for years 1948–2010 (see Fig. D in our response to your last comment), we do not think that the use of the GEOS-4 data compromises the conclusions in our study.

Considering the large uncertainties in chemistry schemes of secondary organic aerosol (SOA), SOA in our simulation is assumed to be the 10% carbon yield of OC from biogenic terpenes (Park et al., 2003) and 2% carbon yield of OC from biogenic isoprene (van Donkelaar et al., 2007; Mu and Liao, 2014). This is now clarified in Sect. 2.2.

Observational studies have shown that SOA accounts for small fractions of OC and $PM_{2.5}$ in winter over the southeastern U.S. (Zheng et al. 2002; Ke et al. 2007; Zheng et al. 2007; Blanchard et al. 2008; Ding et al. 2008; Kleindienst et al., 2007, 2010; Weber, 2010). For example, Kleindienst et al. (2007) showed that SOA contributed about 0.5 μg C m^{-3} (about 18%) to OC in January–February, 2003, by field study carried out at a research site in Research Triangle Park, North Carolina. Weber (2010) compiled SOA observations at 15 sites in the southeastern U.S. in 2007 and showed that SOA concentration averaged over sites was about 0.9 μg m^{-3} (or about 10% of $PM_{2.5}$ mass) due to the low biogenic VOC emissions at low temperatures in winter.

Specific comments.

Abstract. Why do the authors just use observations over 1999- 2003 when data is available from 1999 to 2013?

Response:

Thanks for pointing out this mistake. It has been changed to “1999–2013”.

Section 1. Maybe the author needs to give a literature review using more recent studies.

Response:

We have added about 10 recent studies (published in 2014 and 2015) in our Introduction section.

Section 2.1. Please specify the detrending method.

Response:

We have added the description on detrending method in Sect. 2.1: “Since the observed aerosol concentrations exhibited a significant decreasing trend from 1999 to present in the U.S. due to the reductions in emissions of aerosols and aerosol precursors (Alston et al. 2012, http://www3.epa.gov/airtrends/aqtrends.html#comparison), the long-term linear trend in concentrations is identified by the least-square fit and then removed from the observed concentrations for each site.”

Section 2.2. Please specify if SOA is included in the chemistry model.

Response:

We have added in Sect. 2.2 the description on SOA simulation: “Considering the large uncertainties in chemistry schemes of secondary organic aerosol (SOA), SOA in our simulation is assumed to be the 10% carbon yield of OC from terpenes and and 2% carbon yield of OC from biogenic isoprene (van Donkelaar et al. 2007; Mu and Liao 2014).”

Section 2.3. There are many other definitions of PNA. Can these different definitions affect the conclusion?

Response:

There are three commonly used definitions of PNAI: (1) PNAI defined by Leathers et al. (1991) ($PNAI_{Leathers}$). This is the definition used in our manuscript. (2) PNAI defined by Wallace and Gutzler (1981) ($PNAI_{Wallace}$) as:

$$PNAI_{wallace}=\frac{1}{4}\left[z^{*}(20°N,160°W)-z^{*}(45°N,165°W)+z^{*}(55°N,115°W)-z^{*}(30°N,85°W)\right]$$

, where * denotes the normalized geopotential height at 500 hPa. (3) PANI defined by NOAA Climate Prediction Center (CPC) ($PNAI_{NOAA}$, http://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/month_pna_index 2.shtml). All these three definitions reflect the same atmospheric teleconnections over the region of North Pacific to North America region. The correlation coefficients between $PNAI_{Leathers}$ and $PNAI_{Wallace}$ (or between $PNAI_{Leathers}$ and $PNAI_{NOAA}$) is equal to or larger than 0.94, when we calculate PNAI for 1979–2013 by using the NCEP-2 reanalyzed meteorological data or PNAI for 1986–2006 by using the assimilated GEOS-4 data (see Fig. A below).

The different definitions of PANI do not influence our conclusion. The differences in observed surface-layer aerosol concentrations between the PNA+ and PNA– months on the basis of $PNAI_{Wallace}$ (Fig. B below) and $PNAI_{NOAA}$ (Fig. C below) are similar to those on the basis of $PNAI_{Leathers}$ (Fig. 3 in our manuscript). In Fig. B, the enhancement of $PM_{2.5}$ reached 5–7 μg m^{-3} (or 40–80%) in California, 3–5 μg m^{-3} (40–80%) around the Salt Lake, 3–5 μg m^{-3} (20–40%) over and near the eastern Midwest. Similar results are shown in Fig. C; the enhancement of $PM_{2.5}$ reached 7–9 μg m^{-3} (or 40–80%) in California, 5–7 μg m^{-3} (40–80%) around the Salt Lake, 3–5 μg m^{-3} (20–40%) over and near the eastern Midwest.

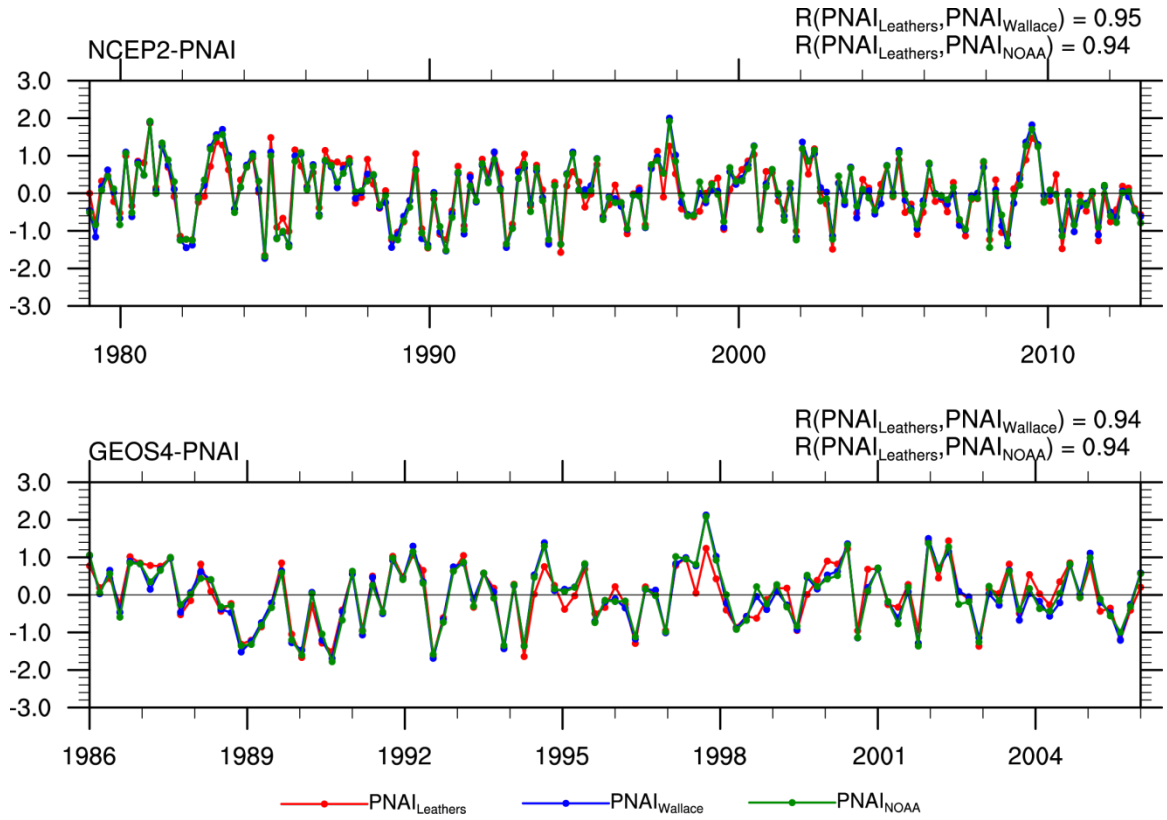

Fig. A. Top panel: Monthly PNAI in NDJFM for years of 1979–2013 calculated using the NCEP-2 data. Bottom panel: PNAI for 1986–2006 calculated using the assimilated GEOS-4 data. The correlation coefficients are marked on top right corner of each panel.

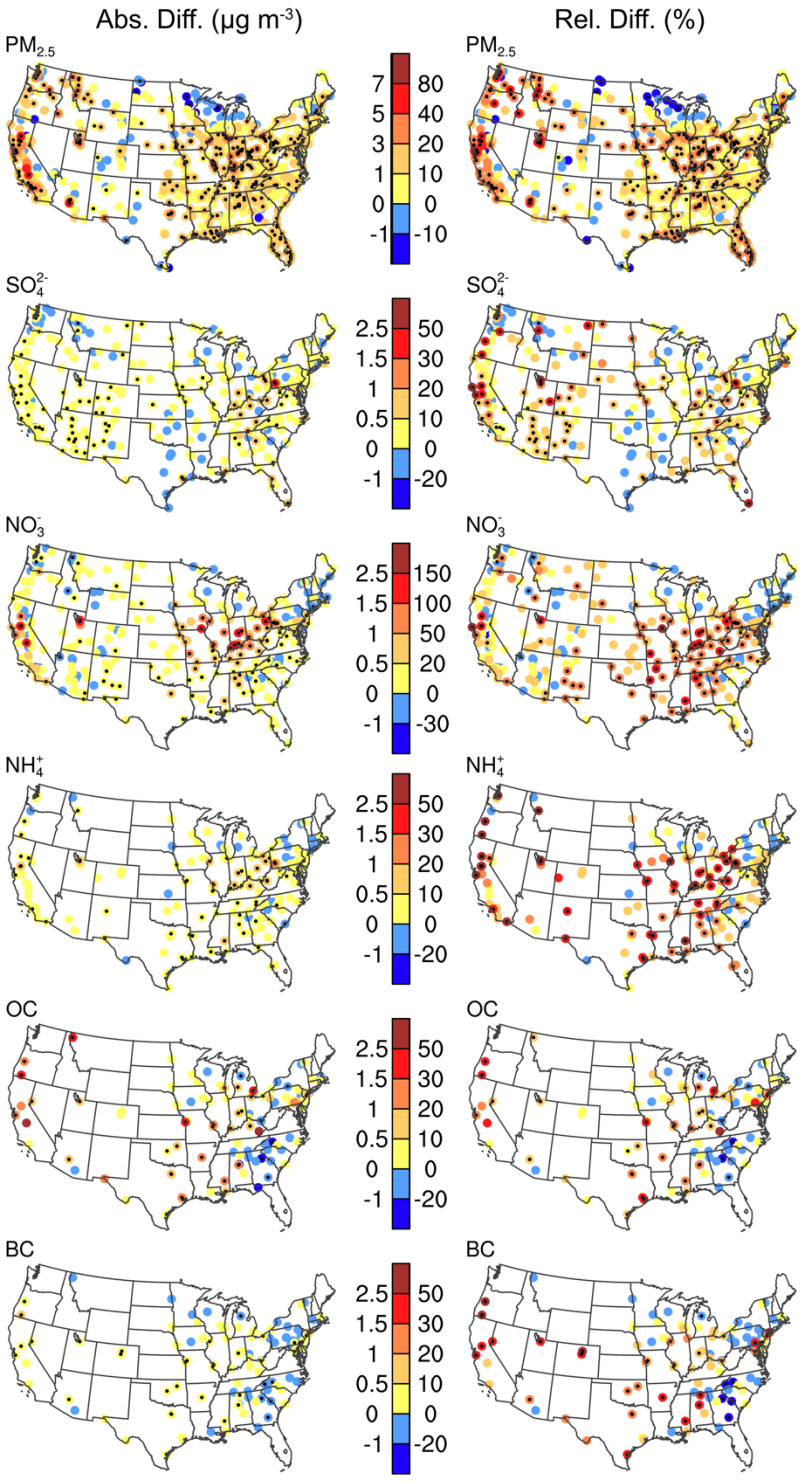

Fig. B. The same as Fig. 3 in the manuscript except that the differences in observed surface-layer aerosol concentrations between the PNA+ and PNA– months are obtained on the basis of $PNAI_{Wallace}$.

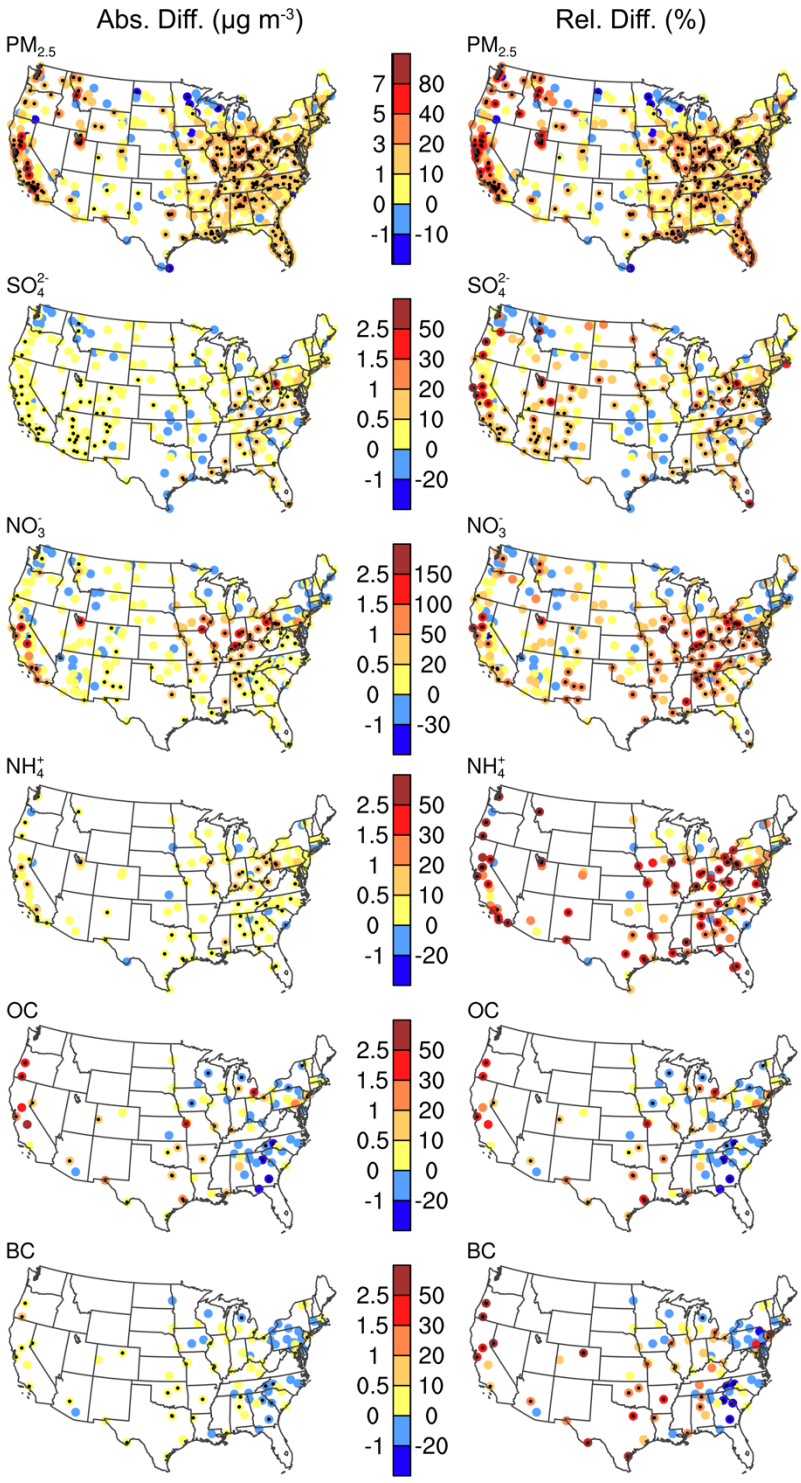

Fig. C. The same as Fig. 3 in the manuscript except that the differences in observed surface-layer aerosol concentrations between the PNA+ and PNA– months are obtained on the basis of $PNAI_{NOAA}$.

Section 3.1. It is very important to include the uncertainty when calculating the PM2.5 concentration difference between positive and negative PNA. And provide details about how you calculate the uncertainty.

Response:

We added the following sentence in Sect. 3.1: “The uncertainty associated with the differences in aerosol concentrations between PNA+ and PNA– months is represented by the two-tail Student-t test with significance level of 90%.”

Section 3.2. The decreasing emission trend should have a large effect on the conclusion about the number of exceedance days. So please show Figure 4 at different timeframes.

Response:

We have removed Sect. 3.2 following the suggestion of Reviewer #3.

Section 4.1. From Figure 5a and 5b, it seems GEOS-Chem largely underestimates the PM2.5 concentrations in California. What is the reason for this? Does it improve if some updated SOA mechanisms is included in the model?

Response:

There are two reasons for the low biases in model results in California. First, the EPA-AQS network had sites in urban and suburban regions, which obtained higher concentrations than other long term networks in the U.S. (Malm et al. 2011; Rattigan et al. 2011; Hand et al. 2012; Hand et al. 2014). Hand et al. (2014) compared the concentrations of aerosols from the EPA-AQS with those from the Interagency Monitoring of Protected Visual Environments (IMPROVE) for 2008–2011, and showed that the ratios of wintertime aerosol concentrations of ammonium sulfate, ammonium nitrate, OC, and BC from the EPA-AQS to those from the IMPROVE were, respectively, 2.3, 7.7, 8.3, and 13.1, as the concentrations were averaged over the western U.S.. Liu et al. (2004) also attributed the high EPA-AQS concentrations in the western U.S. to the relative sparse urban sites that were heavily influenced by strong local sources such as automobiles and wood fires. Second, the low biases in California may also be caused by the systematic biases in emissions in the model. With a high resolution (0.5° latitude by 0.667° longitude over North America) nested-grid version of the GEOS-Chem model (version 9-01-02), Walker et al. (2012) found large underestimates of nitrate and ammonium in California, with NMB of −62% and −38%, respectively, as model results were compared with the IMPROVE and CASTNET observations.

If we update the SOA mechanism, simulated OC concentration in California might be improved slightly but the underestimates in $PM_{2.5}$ in this region cannot be improved, since concentrations of all aerosol species are underestimated in the western U.S.. Heo et al. (2013) showed that the measured SOA was low (about 0.2–0.3 μg m^{-3}) in California in winter, so improving SOA simulation should not have a large impact on simulated OC in California.

Figure 5b shows the spatial correlation between the observations and the GEOS-Chem. How about the temporal correlation in different regions? Maybe the correlation is very low. Can it affect the conclusion?

Response:

Following the reviewer's suggestion, we have calculated the temporal correlation coefficient between EPA-AQS observations and GEOS-Chem model results at each site for each aerosol species ($PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, OC, or BC) (auxiliary Fig. S4, see below). The temporal correlations are statistically significant for $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, at most sites in the U.S., especially over and near the eastern Midwest where largest increases in aerosol concentrations are identified in the PNA+ months relative to the PNA– months. We have added these results of temporal correlation coefficients to the end of Sect. 4.1.

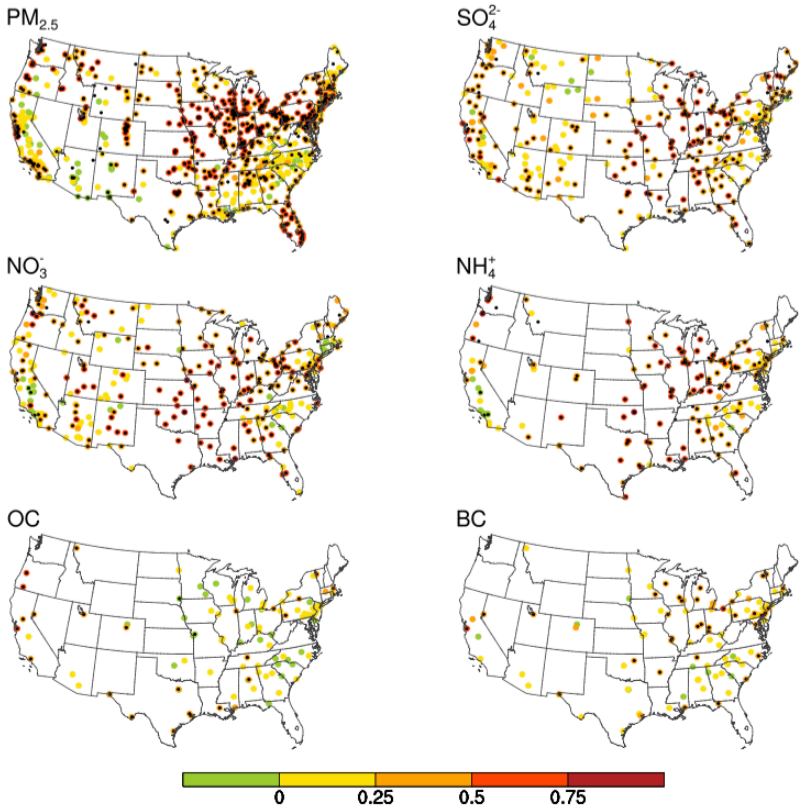

Fig. S4. The temporal correlation coefficient between EPA-AQS observations and GEOS-Chem model results at each site for each aerosol species. Datasets used here are the same as those used in Figure 4 of the manuscript. The sites with black dots are those that have passed the two-tail t-test with 90% confidence level.

Section 5.1 How important is transboundary transport of aerosols? If it contributes to a tiny amount of total PM2.5 mass, this section may be not important.

Response:

The monthly mean $PM_{2.5}$ mass over the selected U.S. box was about 0.10 Tg (integration over the box from the surface to 100 hPa), and the net loss through the boundaries of the box was about 0.32 Tg $month^{-1}$, indicating that the transboundary transport of aerosols is very important.

Also, the authors should discuss whether the underestimate in GEOS-Chem over the west US affects the conclusion in this section.

Response:

We have added a sentence to discuss at the end of Sect. 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 differences in net flux between PNA+ and PNA– phases."

Section 5.2 I would suggest the authors to check the impact of PNA on T, RH, precipitation and surface wind speed using NCEP Reanalysis over a much longer time period, e.g. 1948-2014.

Response:

Following the reviewer's suggestion, we have calculated the impact of PNA on T, RH and surface wind speed over 1948–2014 by using the NCEP Reanalysis and on precipitation over 1948–2010 by using Global Precipitation Climatology Center (GPCC) data (http://www.esrl.noaa.gov/psd/data/gridded/data.gpcc.html) (Fig. D below). The spatial patterns of the differences in meteorological parameters in Fig. D are similar to those shown in Fig. 7 (obtained for years of 1986–2006 by using the GEOS-4 data), except that more grid cells have passed the two-tail student-t test with 90% confidence level in Fig. D with more samples in a longer time period.

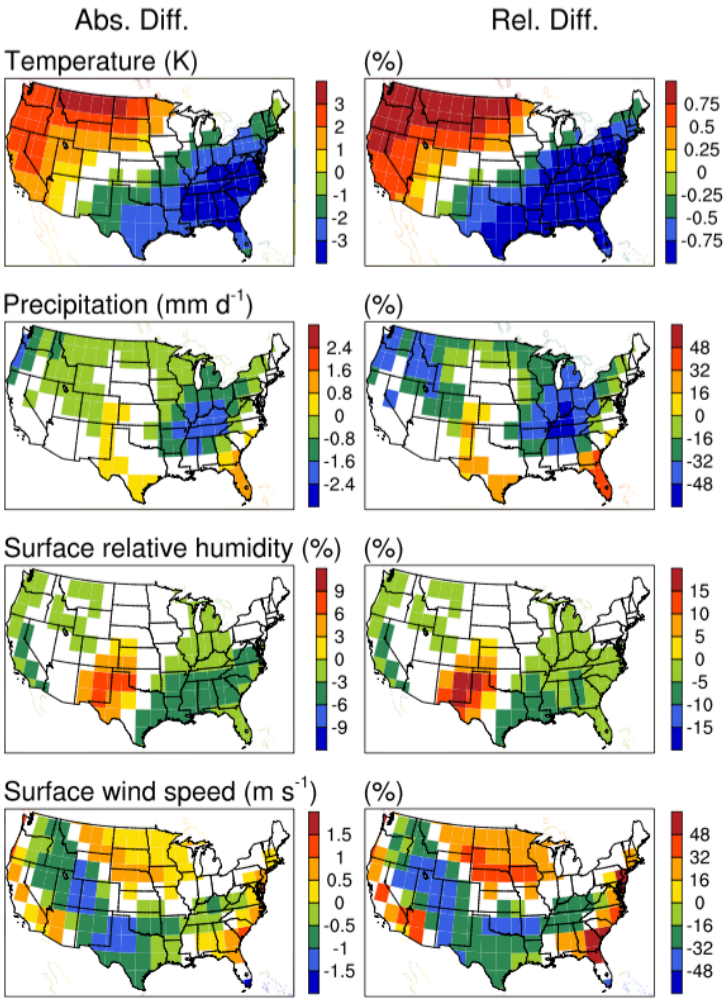

Fig. D. The same as Fig. 7 in the manuscript except that the differences in T, RH, and surface wind speed between PNA+ and PNA– months are calculated for 1948–2014 by using the NCEP Reanalysis and the differences in precipitation are calculated for 1948–2010 by using the Global Precipitation Climatology Center (GPCC) data (http://www.esrl.noaa.gov/psd/data/gridded/data.gpcc.html).

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Manuscript # ACP-2015-804

Responses to Reviewer #2

Overview

This study relates winter variability of particulate matter (PM) to changes in the Pacific-North America climate index. The authors use both observations and model simulations of PM. A statistically significant difference in PM is identified between the positive and negative phases of the PNA, particularly in the US Midwest. The authors provide some statistical evidence that changes in meteorology attributable to the PNA is the cause.

Overall the paper is generally clear. I think the research fits will within the scope of ACP and that the results would be well-received by the community. However, I have a number of concerns. I believe the authors can ease these concerns with additional work. I recommend the manuscript for major revision.

General Comments

I have some concern about the definition of the PNA index. It is not clear why the geopotential height in three grid boxes is sufficient. I recommend using a more standard index. For example, the NOAA Climate Prediction Center provides monthly values of the PNA index using the more common EOF loading approach. Also, just looking by eye, the NOAA CPC PNA data appears to differ from the PNAI presented here.

Response:

There are three commonly used definitions of PNAI: (1) PNAI defined by Leathers et al. (1991) ($PNAI_{Leathers}$). This is the definition used in our manuscript. (2) PNAI defined by Wallace and Gutzler (1981) ($PNAI_{Wallace}$) as: $PNAI_{wallace}=\frac{1}{4}\left[z^*(20°N,160°W)-z^*(45°N,165°W)+z^*(55°N,115°W)-z^*(30°N,85°W)\right]$, where * denotes the normalized geopotential height at 500 hPa. (3) PANI defined by NOAA Climate Prediction Center (CPC) ($PNAI_{NOAA}$, http://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/month_pna_index 2.shtml). All these three definitions reflect the same atmospheric teleconnections over the region of North Pacific to North America region. The correlation coefficients between $PNAI_{Leathers}$ and $PNAI_{Wallace}$ (or between $PNAI_{Leathers}$ and $PNAI_{NOAA}$) is equal to or larger than 0.94, when we calculate PNAI for 1979–2013 by using the NCEP-2 reanalyzed meteorological data or PNAI for 1986–2006 by using the assimilated GEOS-4 data (see Fig. A below). Therefore it is sufficient to use $PNAI_{Leathers}$ in our study.

By using the PNAI defined by geopotential heights in 3 grid boxes ($PNAI_{Leathers}$) and in 4 grid boxes ($PNAI_{Wallace}$), many previous studies examined the variation and mechanism of PNA as well as the relationship between PNA and surface weather over the U.S. (Yarnal and Diaz, 1986; Carleton et al., 1990; Leathers et al., 1991; Leathers and Palecki, 1992;

Trenberth and Hurrell, 1994; Rodionov and Assel, 2001; Schoof and Pryor, 2006).

There are three reasons for the differences between PNAI shown in our Fig. 2 and that shown by NOAA CPC (http://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/month_pna_index.shtml). First, the calculation depends strongly on the length of samples. The NOAA CPC PANI has the samples covering all months throughout the year over 1950–present, with the mean value and variance of the time series different from what we have in our calculation of PNAI (in our work we are only concerned with the PNAI for the months of NDJFM over 1986–2013). Second, the NOAA CPC PANI has been smoothed by a 3-month running mean method. Third, the NOAA CPC PANI is calculated by using the geopotential height with seasonal cycle, but the PNAI is our work is calculated by using the deseasonalized geopotential height to isolate the monthly variation.

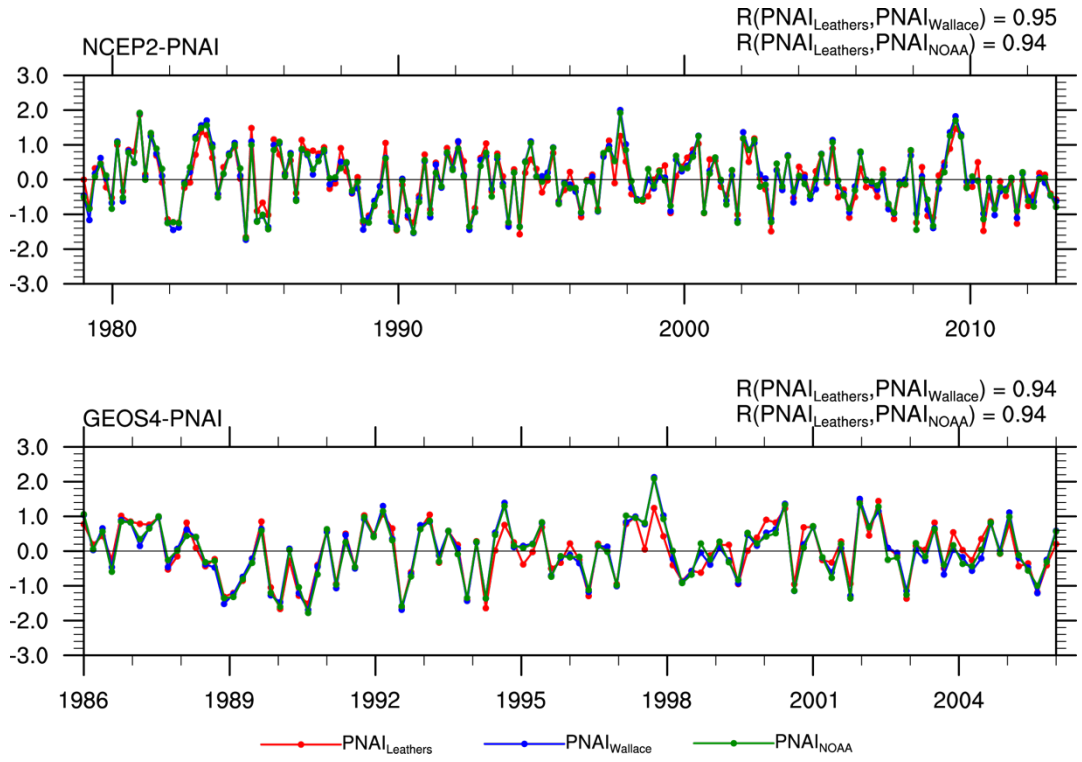

Fig. A. Top panel: Monthly PNAI in NDJFM for years of 1979–2013 calculated using the NCEP-2 data. Bottom panel: PNAI for 1986–2006 calculated using the assimilated GEOS-4 data. The correlation coefficients are marked on top right corner of each panel.

- Most of the analysis focuses on differences between the positive and negative phases of the PNA. It is generally most constructive to isolate the impacts of each phase from neutral conditions. If the authors have motivation to compare positive vs. negative phases, they should include those thoughts in the manuscript.

Response:

The method of compositing the differences between the positive and negative phases of one atmospheric circulation index (ACI) is a commonly

used approach to analyze the impact of the ACI on a specific phenomenon. Most importantly, the composite differences can show the magnitude of the influence of ACI. Such approach has been used in many studies to examine the impact of climate variability on concentrations of aerosols (Di Pierro et al., 2011; Singh and Palazoglu, 2012; Zhu et al., 2012; Jerez et al., 2013; Qu et al., 2015) and on concentrations of tracer gases (Eckhardt et al., 2003; Liang et al., 2005; Jerez et al., 2013; Yang et al., 2014).

- The definition of the PNAI appears to have a significant flaw. I think this may just be a mistake in how the equation is written. By my calculation the denominator of Z*'_i,j is always 0. The summation can be disturbed across the parentheses (since the terms are simply subtracted). The first term generates the average value of Z' (which should be 0 anyway, see below). Summing the second term is trivial since there is no dependence on i or j, so it is multiplied by Nx5 and then divided by Nx5. Thus both terms give the average value of Z': Z'-Z'=0.

The second term of the numerator, by my calculation, should also be 0. The summation of anomalies from a mean will always be equal to 0, if the period used for estimation of the mean and period of anomalies are the same.

This equation needs correction; it is clearly not the algorithm used to produce the data in Fig. 2. Perhaps this is an issue with notation. See additional comment about these equations below.

Response:

Thanks for pointing out the typographic errors. We now have the following corrected equation in the revised manuscript:

$$Z^{*'}_{i,j} = \frac{Z'_{i,j}}{\sqrt{\frac{1}{n\times 5}\sum_{i=1}^{n}\sum_{j=1}^{5} Z'^{\,2}_{i,j}}}$$

- There are significant trends in PM2.5 and aerosol composition over this time period (hints of thus can be seen in Fig. 5c). The EPA AQS data should be detrended (in addition to removing seasonality) in order to best detect interannual variability. It needs to be shown that differences arise from true variability and not other factors (emission regulations). Indeed, just by eye, most of the positive phase months are in the early part of the period and negative at the end. This could introduce an artifact of emission regulations.

Response:

This is a good point. We have redone the composite analyses for observed aerosol concentrations by detrending the observations from the EPA-AQS. The revised differences in aerosol concentrations between positive and negative PNA are similar to the results in our previous version of manuscript (those obtained without detrending); the horizontal distributions are about the same but the magnitudes of the differences in aerosol concentrations are slightly smaller. We have updated Figs. 3, 4 and Table 1, and have changed the descriptions in the text accordingly.

- How are regional averages performed? A US-scale average will generally be more heavily weighted by the East Coast since there are more sites in that region. Some of the regional average numbers presented (US, western US, eastern US) should be reassessed.

Response:
We agree with the Reviewer. We have added a sentence in Sect. 3 to clarify this: "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."

- The figures are very hard to view. I recommend breaking up some of them to allow larger maps. Figures 3, 5, and 6 are particularly hard to read. The data in the maps is useful and necessary.

Response:
The original figures are not so small. The current very small figures were caused by the landscape pages of ACPD. It is expected that the figures will be easier to view when each of these figures can take up a whole portrait page in its ACP version.

Specific Comments - Page 33210, Line 6: "...Air Quality System of Environ- mental . . ." should be ". . .Air Quality System of the Environmental. . ."

Response:
Changed.

- Page 33215, Line 4: It is unclear if 5 days of observations are required or 5 observational periods. A single observational period could be 5 days long.

Response:
We have clarified this as "… there were at least 5 observation records within each month."

- Page 33217, Line 14: The notation here is confusing. The equation defining Z' should not have two references to i (i is listed as an input and the summation variable).

Response:
Corrected.

- Page 33218, Line 2: "PANI" should be "PNAI".

Response:
Changed.

- Page 33218, Line18: I do think contiguous Salt Lake is an identifiable location.

Response:

We have changed “the contiguous Salt Lake” to “the contiguous Salt Lake (northern Utah)”.

- Page 33219, Line 9: Why is 90th percentile used? 95th percentile is more standard.

Response:

The 90% confidence level for t-test is also commonly used in previous studies to examine the impacts of atmospheric circulation on aerosols (Schultz et al., 2007; Wu et al., 2008, 2013; Gong et al., 2010; Hirdman et al., 2010; Qian et al., 2011; Jerez et al., 2013; Huang et al., 2014; Gettelman et al., 2015).

- Figure 2: The difference between Figure 2a and 2b is not clear. The panels have slightly different labels, but the data and highlighted +/- points appear identical.

Response:

The Fig. 2a (Fig. 2b) was the PNAI for analyzing observed $PM_{2.5}$ (individual aerosol species) over 1999–2013 (2000–2013). Figs. 2a and 2b are almost identical for the highlighted +/– points except for those in year 1999. See Sect. 2.3 for our definitions of PNA+ and PNA–.

- Page 33222, Line 23: Again, you cannot use the variable i in the summation notation since its already used to denote the month of interest.

Response:

We have changed *i* to *m*, *n* to *M*, and the revised equation is:

$$DM_m=(C_m - \frac{1}{M}\sum_{m=1}^{M} C_m)/\frac{1}{M}\sum_{m=1}^{M} C_m$$

- Table 3 – The mass flux values seem a little low. I recommend double-checking the calculation.

Response:

We have checked our calculation and the results are right.

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Manuscript # ACP-2015-804

Responses to Reviewer #3

This manuscript examines the correlation between the Pacific-North America (PNA) phases and the wintertime surface-layer aerosol concentrations at the United States (US) on monthly scale. It uses the observation (1999-2013) from the Air Quality System of Environmental Protection Agency (EPA-AQS) and the simulation (1986-2006) from Goddard Earth Observing System chemical transport model (GEOS-Chem). Both the observation and simulation are composite to months with positive and negative PNA phases. The mean aerosol loadings are found to be higher for PNA+ months. The authors tried to understand the mechanisms for the PNA impacts on aerosol concentrations with the chemical transport model (CTM). Cross-boundary fluxes are found to be responsible for the PM2.5 increase in PNA+ months. With the pattern correlation coefficients (PCCs), they quantified the relative influence on local aerosol abundances from temperature, precipitation, relative humidity, wind, and boundary layer height.

The manuscript is well organized, and within the scope of ACP. Overall, I found that the observation-based analysis on the PNA impact on aerosol was more valuable than the model-based attempt on understanding the driving mechanisms. It can be published after addressing the following questions/comments.

1. Although it is mentioned that chemical reactions affect the aerosol concentrations, it seems assumed that large-scale circulation and/or meteorological factors dominate the chemical factors. Some references showing the relative contributions of dynamics, physics, and chemical impacts on aerosols are helpful.

Response:

The meteorological parameters influence the concentrations of aerosols by both physical and chemical processes. The second paragraph of our introduction section reviews mainly the impacts of meteorological parameters (such as temperature and relative humidity) on chemical reactions, and the third paragraph of this section is focused on the impacts of atmospheric circulation on aerosols. Since the impacts of meteorological parameters on physical and chemical processes are coupled, few studies have quantified the relative contributions of dynamics, physics, and chemical impacts.

2. It is known for years that the GEOS-Chem model has high biases in OH (chemistry is too “hot”, resulting for example too short methane lifetime). Is this problem improved in the GEOS-Chem version used here? What is the CH4 lifetime in the model? The oxidizing capacity of the troposphere is largely determined by OH, which is a chemical sink for the aerosols studied here. It is

hard to represent aerosols correctly in the model without reasonable OH, unless the chemical impact is trivial. In fact, the biases in the chemistry simulation may explain some of the model-obs differences denoted in Sect. 4.1.

Response:

Wu et al. (2007) simulated methane lifetime by using the GEOS-Chem version 7-04-02 and reported a methane lifetime of 10.3 yr. Holmes et al. (2013) examined methane lifetime on the basis of the GEOS-Chem (version 9-01-02) simulation and reported a lifetime of 9.4–10.2 yr. Therefore, methane lifetime from the GEOS-Chem agrees closely with the lifetime of 11.2±1.3 yr constrained by methyl chloroform observations by Prather et al. (2012). For the version 8-02-01 of the GEOS-Chem model used in our study, the 1 yr benchmark run obtained an annual and tropospheric mean OH concentrations of 1.18×10^{6} molec cm^{-3} (http://wiki.seas.harvard.edu/geos-chem/index.php/Mean_OH_concentration), which is less than the OH concentration (1.22×10^{6} molec cm^{-3}) from the version 9-01-02 by Holmes et al. (2013) and the OH concentration (1.24×10^{6} molec cm^{-3}) from version 7-04-02 by Wu et al. (2007).

The aerosol concentrations are not so sensitive to OH concentrations in model simulations. Rae et al. (2007) examined the sensitivity of sulfate to oxidant concentration and climate, and found that the global sulfate burden would decrease by about 3% with an 11.7% reduction in OH burden over 1990–2100. However, sulfate burden would increase by 9% owing to climate change alone over 1990–2100. Heald et al. (2012) found that a reduction in OH levels by 25% did not influence the simulated nitrate levels (within 5% of the baseline concentrations), because a reduction in nitric acid formation was somewhat compensated by an increase in lifetime. Hence the biases in simulated OH concentrations should not the key reason for the model-obs differences presented in Sect. 4.1.

2. Sect. 5.2 uses the PCC to quantify the relative contribution of different meteorological variables to the aerosol changes due to different PNA phases. It finds some correlations. But more sophisticated analysis is required to infer the driving mechanisms.

Response:

These are three commonly used methods to quantify the relative contribution of different meteorological variables: the multi linear regression (MLR) (Tai et al, 2010, 2012), the sensitivity experiments (SE) (Aw and Kleeman, 2003; Dawson et al, 2007; Kleeman, 2007), and the pattern correlation coefficients (PCC) (Levy et al., 2008; Jeong and Park, 2013; Allen et al., 2015). MLR quantifies the relative contribution of different meteorological variables by establishing the temporal empirical relationship between aerosol concentrations and the meteorological variables, but it does not concern with the spatial distribution of the influence of meteorological variables. SE, by changing one specific meteorological variable (for example, PBLH) ignores the covariance of other meteorological parameters (such as temperature and convection associated with PBLH), which cannot give us a realistic responses of aerosol concentrations to changes in meteorological

conditions. PCC quantifies the relationship between meteorology and aerosol concentrations both spatially and temporally.

We use the magnitude of the pattern correlation coefficients (PCC) to quantify the relative contribution of different meteorological variables to the changes in aerosols. The same statistical approach was also used in previous studies (Levy et al., 2008; Jeong and Park, 2013; Allen et al., 2015). Levy et al. (2008), by simulations with the NOAA/GFDL climate model, reported that the regional pattern of changes in surface temperature did not correspond well to the regional pattern of radiative forcings of short-lived species because of the low global PCC of –0.172 under the IPCC A1B scenario. Jeong and Park (2013) used the PCC values to show the impacts of variabilities in temperature, precipitation, relative humidity, mixing depth, cloud fraction, and surface wind speed on regional patterns of concentrations of O_3, $(NH_4)_2SO_4$, and NH_4NO_3 in East Asia on the basis of the GEOS-Chem simulation. Allen et al. (2015) quantified the role of precipitation in the changes of aerosol burdens from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP); they obtained global PCC values of –0.36, –0.33 and –0.30 between precipitation and burdens of SO_4^{2-}, BC, and OC, respectively.

Specific comments:

1. Sect. 2.2, besides noting the methane (or other species that can infer OH concentration) lifetime, it is worth mentioning how many layers are in the PBL as the focus of the study is the aerosols in the surface layer, which is largely impacted by the presentation of PBL structure in the model.

Response:

We have added the following sentence in Sect. 2.2: “The instantaneous vertical mixing in the planetary boundary layer (PBL) is accounted for by the TURBDAY mixing scheme (Bey et al., 2001). The PBL occupies the lowest 3–6 vertical model layers over the U.S..”

2. P33218, L24, what are the numbers if restrict to the sites that pass the t test? Are they significantly different from current ones? Try to get a sense on how different sampling changes the results.

Response:

Following the suggestion, we have recalculated the average differences between PNA+ and PNA– phases by using the sites that pass the t test. The results are shown in Table A (see below). The differences were larger than those calculated with the sites that both pass and do not pass the t-test with 90% confidence level.

Table A. The absolute (μg m^{-3}) and relative (%) differences in observed aerosol concentrations between the PNA+ and PNA– months (PNA+ minus PNA–). The observed concentrations are averaged over the sites that have passed the two-tail student-t test with 90% confidence level in the whole of U.S., in the western U.S., or in the eastern U.S.. 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
$PM_{2.5}$	1.9 (17.6%)**	2.7 (30.5%)**	1.7 (14.9%)**
SO_4^{2-}	0.1 (6.3%)	0.2 (20.3%)**	0.2 (5.4%)
NO_3^-	0.6 (68.8%)**	0.5 (67.2%)**	0.7 (71.6%)**
NH_4^+	0.3 (27.2%)*	0.5 (72.0%)**	0.3 (22.7%)**
OC	0.5 (11.3%)**	0.6 (13.0%)**	0.4 (10.5%)**
BC	0.3 (44.3%)**	0.3 (39.3%)**	0.3 (47.4%)**

3. Sect. 3.2 can be removed. It is very short and does not add much, if any, to the paper. The results shown in Fig. 4 are mostly numbers close to 0. No significance test is done for those numbers, so I am not sure statistically how significant those numbers are. If the authors would like to keep this section, I'd highly recommend the significance test. The tools used for extreme weather/climate can be borrowed, which often rely on shifted (towards high tail) probability distribution functions.

Response:

We have removed Sect. 3.2 following your suggestion.

4. P33222, L10-18: How much of the underestimated modeling PM2.5, SO4, NO3, etc. is caused by "too hot" chemistry in the model as mentioned above? The authors seem believe that it is mainly because of the observational uncertainties. Even it is the case, a factor of up to 13 differences in different measurements is worrisome. Such large error bars of the observations bring up the question if those observations are suitable for scientific applications.

Response:

We think that the biases in simulated OH concentrations should not the key reason for the model-obs differences (see our response to #2 of your general comments).

There are two reasons for the low biases in model results. First, the EPA-AQS network had sites in urban and suburban regions, which obtained higher concentrations than other long term networks in the U.S. (Malm et al. 2011; Rattigan et al. 2011; Hand et al. 2012; Hand et al. 2014). Hand et al. (2014) compared the concentrations of aerosols from the EPA-AQS with those from the Interagency Monitoring of Protected Visual Environments (IMPROVE) for 2008–2011, and showed that the ratios of wintertime aerosol concentrations of ammonium sulfate, ammonium nitrate, OC, and BC from the EPA-AQS to those from the IMPROVE were, respectively, 2.3, 7.7, 8.3, and 13.1, as the concentrations were averaged over the western U.S.. Liu et al. (2004) also attributed the high EPA-AQS concentrations in the western U.S. to the relative sparse urban sites that were heavily influenced by strong local sources such as automobiles and wood fires. Second, the low biases may also be caused by the systematic biases in emissions in the model. With a high resolution (0.5° latitude by 0.667° longitude over North America) nested-grid version of the GEOS-Chem model (version 9-01-02), Walker et al. (2012) found large underestimates of nitrate and ammonium in California, with NMB

of −62% and −38%, respectively, as model results were compared with the IMPROVE and CASTNET observations.

The EPA observations together with the GEOS-Chem simulation were used by previous studies (for example, Liu et al., 2004; van Donkelaar et al., 2006; Drury et al., 2010; Tai et al., 2010, 2012). Even though there were biases in EPA observations, the statistically significant differences in aerosol concentrations between PNA+ and PNA– months (Fig. 3 of our manuscript) still provide valuable information for the purpose of our study.

5. P33223, L5-10: I understand why the GFED OC and BC lines are added to those plots. I suggest adding one or two sentences explaining the reason for it, so it would read logically smoother.

Response:

We have added a sentence here: “Since the biomass burning emissions, which contribute largely to carbonaceous aerosols, have large interannual variations (Duncan et al., 2003; Generoso et al., 2003; van der Werf et al., 2006), we also show in Fig. 4c the time series of DMs of biomass burning emissions of OC and BC by using biomass burning emissions in NDJFM over 2000–2006 from GFED v2.”

6. Sect. 4.2: Figures 3 and 6 are compared extensively. It would be better if the corresponding panels (abs and relative differences) use the same color bars. Otherwise, it is difficult to see some of the features noted in the text.

Response:

Thanks for the suggestion. We now use the same color bars for relative differences in Figs. 3 and 6 of the revised manuscript. We have tried to use same color bars for absolute differences but decided to keep using different color bars in these two figures. Otherwise it will be difficult to see spatial distributions of the absolute differences between PNA+ and PNA– phases due to the low biases in simulated concentrations.

7. Table 4: There is no point to show the numbers that fail the significance test. I’d suggest changing them to NaNs or dashes.

Response:

To be consistent with Tables 1 and 2, we think it is better to keep the PCC numbers passed or failed the significant test in Table 3. All correlation coefficients were presented in the studies of effects of meteorology or atmospheric circulations on pollutants (Gong et al., 2006; Liang et al., 2005; Vukovich, 1995; Liu et al., 2013; Wu et al., 2013).

8. Figure 2: It is redundant showing panels (a) and (b) only because different years of PNA phases are marked. I suggest that either (a) or (b) be deleted, but marking the PNA+/- months for the whole period (1986-2013). It will allow comparing the NCEP2- PNAI and GEOS4-PNAI for more years than the current form.

Response:

The PNAI are shown in those panels because of our definition of the PNAI phases: the positive PNA months (PNA+) were 25% of the $n\times5$ PNAI months with the highest positive PNAI values, the negative PNA months (PNA–) were 25% of the $n\times5$ PNAI months with the highest negative PNAI values, and the rest months were referred to as the transitional months (see Sect. 2.3). The Fig. 2a (Fig. 2b) was the PNAI for analyzing observed $PM_{2.5}$ (individual aerosol species) over 1999–2013 (2000–2013). Figs. 2a and 2b are almost identical for the highlighted +/– points except for those in year 1999.

9. Figure 3: The caption should mention the results are for monthly averages (it says in the text, but should be clear in the caption as well). Also, the test is to confirm if the two averages (PNA+/-) are significantly different. The caption should be revised to be more specific about this.

Response:

We have revised the figure caption as: "The absolute ($\mu g\ m^{-3}$, left column) and relative differences (%, right column) in observed monthly mean aerosol concentrations between PNA+ and PNA– months (PNA+ minus PNA–). 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. 2a. The measurements of speciated aerosols were taken during 2000–2013, in which there were 17 PNA+ and 17 PNA– months (Fig. 2b). The sites with black dots were the differences that passed the two-tail t-test with 90% confidence level."

10. Figure 5(b): Many panels show clearly two regimes. Does it make sense to fit a single line? Maybe two lines are better?

Response:

Following the suggestion, we now show fits for the eastern, western, and whole of U.S., as represented by red, blue, and black lines, respectively.

11. Figure 5 caption: Space is missing "DMof"

Response:

Corrected.

Reference:

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

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Abstract

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 wintertime. This study examined the impacts of monthly variation of the PNA phase (positive or negative phase) on wintertime surface-layer aerosol concentrations in the U.S. by analyzing observations during 1999–2013 from the Air Quality System of the Environmental Protection Agency (EPA-AQS) and the model results for 1986–2006 from the global three-dimensional Goddard Earth Observing System (GEOS) chemical transport model (GEOS-Chem). The composite analyses on the EPA-AQS observations over 1999–2013 showed that the average concentrations of $PM_{2.5}$, sulfate, nitrate, ammonium, organic carbon, and black carbon aerosols over the U.S. were higher in the PNA positive phases than in the PNA negative phases by 1.0 μg m^{-3} (8.7%), 0.01 μg 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 m^{-3} (27.8%), respectively. The simulated geographical patterns of the differences in concentrations of all aerosol species between the PNA positive and negative phases were similar to observations. Based on the GEOS-Chem simulation driven by the assimilated meteorological fields, the PNA-induced variation in planetary boundary layer height was found to be the most dominant meteorological factor that influenced the concentrations of $PM_{2.5}$, sulfate, ammonium, organic carbon, and black carbon, and the PNA-induced variation in temperature was the most important parameter that influenced nitrate aerosol. Results from this work have important implications for understanding and prediction of air quality in the United States.

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.

Previous studies have shown that aerosol concentrations are very sensitive to meteorological parameters (Aw and Kleeman 2003; Wise and Comrie 2005; Dawson et al. 2007; Kleeman 2008; Jacob and Winner 2009; Tai et al. 2010; Tai et al. 2012a; Allen et al. 2015; Markakis et al. 2015; Megaritis et al. 2014; Porter et al. 2015). Aw and Kleeman (2003) examined the sensitivity of $PM_{2.5}$ (aerosol particles which diameter ≤ 2.5 μm) concentration to temperature by performing sensitivity studies in the California Institute of Technology/UC Davis (CIT/UCD) air quality model. A 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 μg m^{-3} (~30%). Wise and Comrie (2005) reported, by statistical analyses of observational datasets obtained from Air Quality System of U.S. Environmental Protection Agency (EPA-AQS), that the variations in meteorological parameters accounted for 20–50% of the variability in aerosol levels over 1990–2003 in five metropolitan areas in the southwestern U.S.. They found that aerosols in these five cities were most sensitive to relative humidity. Dawson et al. (2007) found, by sensitivity studies in the Particulate Matter Comprehensive Air Quality Model with extensions (PMCAMx), that $PM_{2.5}$ concentrations in summer had a small sensitivity to temperature increases (−16 ng m^{-3} K^{-1} on average) because the increases in sulfate offset the decreases in

74 nitrate and organics, while $PM_{2.5}$ concentrations in winter decreased significantly with
75 temperature (−170 ng m^{-3} K^{-1} on average) because the increases in temperature led
76 to large reductions in nitrate and organics. Dawson et al. (2007) also showed that
77 $PM_{2.5}$ concentrations increased with humidity in both winter and summer. Jacob and
78 Winner (2009) summarized by literature review that the regional stagnation, mixing
79 depth, and precipitation are the most important meteorological parameters that
80 influence surface-layer aerosol concentrations. Future climate change was also
81 simulated to influence aerosol levels over the U.S. by about 1 μg m^{-3} (Jacob and
82 Winner 2009), as a result of the climate-induced changes in atmospheric oxidants,
83 transport, deposition, and the shift of gas-particle equilibria (Liao et al. 2006; Unger et
84 al. 2006; Bauer et al. 2007; Jacob and Winner 2009; Pye et al. 2009; Lam et al. 2011;
85 Day and Pandis 2011; Juda-Rezler et al. 2012; Tai et al. 2012b).

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

depth (AOD) of dust in summer correlated with the NAO index during 1983–1994 with a correlation coefficient of 0.50. Jerez et al. (2013) found, by simulations of aerosols for years 1970–1999 with the CHIMERE chemistry transport model driven by the BCMWF ERA40 reanalysis data, that the concentrations of PM_{10} and $PM_{2.5}$ in the southern European regions in winter differed by 10 and 20 μg m^{-3}, respectively, between the positive and negative NAO phases. By using the multi-angle imaging spectroradiometer satellite (MISR) datasets of AOD during 2000–2011, Liu et al. (2013) found a period of 3–4 years in observed summertime AOD over the North China Plain (NCP), and the peak of summertime AOD in NCP occurred four months later after the rapidly transition of El Nino from a warm phase to a cold phase because of the associated cyclone anomaly and maritime inflow over the NCP. Singh and Palazoglu (2012) found correlations between PDO and ENSO and the aerosol exceedance days (defined as the days with $PM_{2.5}$ concentrations larger than the U.S. National Ambient Air Quality Standard) at 6 regions in the U.S. by using the EPA-AQS $PM_{2.5}$ wintertime datasets during 1950–2008.

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

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

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

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

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 aerosol concentrations during 1999–2013 from EPA-AQS and also by simulations of aerosol concentrations for years 1986–2006 using the global chemical transport model GEOS-Chem. The scientific goals of this work are (1) to quantify the differences in wintertime concentrations of sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), black carbon (BC), organic carbon (OC), and $PM_{2.5}$ in the U.S. between different PNA phases, and (2) to understand the roles of PNA-induced variations in meteorology (for example, surface air temperature, wind speed, planetary boundary layer height, precipitation, and relative humidity) in influencing the wintertime aerosol concentrations. The definition of the PNA index, the EPA-AQS observation data used 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 concentrations in the U.S. obtained from the EPA-AQS observations and the GEOS-Chem simulation, respectively. The mechanisms for the impacts of PNA on aerosols are examined in Sect. 5.

2 Data, simulation, and methodology

2.1 Observed aerosol concentrations

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

The measurements of aerosol concentrations from the EPA-AQS were carried out at various time intervals (for example, with measurements every one, three or six days) at different sites, and there were plenty of missing values at many sites. The observed concentrations are pre-processed following the three steps: (1) For a specific site, the observations are used in our analyses if the site had at least 5 months of observations and there were at least 5 observation records within each month. (2) The mean seasonal cycle in aerosol concentrations in the months of November-March is removed by using the similar approach to that used for PNA index (see Sect. 2.3 below). Such deseasonality approach was used in previous studies that examined the monthly variations in mineral dust aerosol (Cakmur et al. 2001; Mahowald et al. 2003), the decreasing trends in observed $PM_{2.5}$ concentrations and satellite AOD in the southeastern U.S. over 2000–2009 (Alston et al. 2012), and the monthly variations in global AOD (Li et al. 2013). (3) Since the observed aerosol concentrations exhibited a significant decreasing trend from 1999 to present in the U.S. due to the reductions in emissions of aerosols and aerosol precursors (Alston et al. 2012, http://www3.epa.gov/airtrends/aqtrends.html#comparison), the long-term linear trend in concentrations is identified by the least-square fit and then removed from the observed concentrations for each site. The EPA-AQS sites with measurements that meet the criteria described in (1) are shown in Fig. 1.

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 assimilated meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). The version of the

199 model we use has a horizontal resolution of 2° ×2.5° and 30 hybrid sigma-P layers
200 from the surface to 0.01 hPa altitude. The model has a fully coupled simulation of
201 tropospheric O_3-NO_x-VOC chemistry and aerosols including SO_4^{2-}, NO_3^-, NH_4^+, BC,
202 OC (Park et al. 2003; Park et al. 2004), mineral dust (Fairlie et al. 2007), and sea salt
203 (Alexander et al. 2005). Considering the large uncertainties in chemistry schemes of
204 secondary organic aerosol (SOA), SOA in our simulation is assumed to be the 10%
205 carbon yield of OC from biogenic terpenes (Park et al., 2003) and 2% carbon yield of
206 OC from biogenic isoprene (van Donkelaar et al., 2007; Mu and Liao, 2014). We
207 mainly examine simulated anthropogenic aerosols from the GEOS-Chem simulation,
208 since mineral dust concentrations in winter are very small (Malm et al. 2004; Zhang et
209 al. 2013) and sea salt is not a major aerosol species in the U.S. (Malm et al. 2004).
210 The model uses the advection scheme of Lin and Rood (1996), the deep
211 convective scheme of Zhang and McFarlane (1995), the shallow convection scheme
212 of Hack (1994), the wet deposition scheme of Liu et al. (2001), and the dry deposition
213 scheme of Wesely (1989) and Wang et al. (1998). The instantaneous vertical mixing
214 in the planetary boundary layer (PBL) is accounted for by the TURBDAY mixing
215 scheme (Bey et al., 2001). The PBL occupies the lowest 3–6 vertical model layers
216 over the U.S..
217 We simulate aerosols for years of 1986–2006 driven by the GEOS-4 reanalysis
218 data. The years of 1986–2006 are chosen for chemistry-aerosol simulation because
219 these are the years that the GEOS-4 datasets are available. Global anthropogenic
220 emissions are from the Global Emissions Inventory Activity (GEIA) (Park et al. 2004;
221 Park et al. 2006; Zhu et al. 2012; Yang et al. 2015). Anthropogenic emissions over the
222 U.S. are overwritten by the U.S. EPA National Emission Inventory for 1999 (NEI99),
223 which have monthly variations in emissions of precursors including SO_2, NO_x, and

224 NH_3. Monthly biomass burning emissions are taken from the Global Fire Emissions
225 Database version 2 (GFED-2) (Giglio et al. 2006; van der Werf et al. 2006). During
226 the simulation of aerosols for years of 1986–2006, the global anthropogenic and
227 biomass burning emissions of aerosols and aerosol precursors are fixed at year 2005
228 levels, so that the variations in aerosol concentrations are caused by variations in
229 meteorological parameters (PNA phases) alone.

230 Natural emissions of O_3 precursors, including biogenic NMVOCs and NO_x from
231 lighting and soil, are allowed to vary over 1986–2006 following the variations in the
232 GEOS-4 meteorological parameters. Biogenic NMVOC emissions are calculated
233 using the module of Model of Emissions of Gases and Aerosols from Nature
234 (Guenther et al. 2006). Lightning NO_x emissions are described by Sauvage et al.
235 (2007) and Murray et al. (2012). Soil NO_x emissions are calculated using the
236 algorithm proposed by Yienger and Levy (1995).

237 The GEOS-Chem simulation of aerosols in the U.S. have been evaluated
238 extensively by previous studies (Park et al. 2003; Park et al. 2004; Park et al. 2005;
239 Heald et al. 2006; van Donkelaar et al. 2006; Liao et al. 2007; Heald et al. 2008; van
240 Donkelaar et al. 2008; Fu et al. 2009; Drury et al. 2010; Leibensperger et al. 2011;
241 Zhang et al. 2012). These studies have shown that the GEOS-Chem model can
242 capture the magnitudes and distributions of aerosols in the U.S..

243 **2.3 PNA index**

244 The PNA index (PNAI) is commonly used to quantify the changes in PNA phase
245 (Wallace and Gutzler 1981; Leathers et al. 1991). This study follows the definition of
246 PNAI by Leathers et al. (1991). In order to examine the monthly variations in PNA, the
247 mean seasonal cycle of geopotential height at 700 hPa is removed for the months of
248 November, December, January, February, and March (NDJFM) in the studied years.

Such deseasonality approach has been used in the analyses of the growth and decay of PNA phase in NDJFM (Feldstein 2002), the development of NAO (Feldstein 2003), the influence of NAO on precipitation in Europe (Qian et al. 2000), and the variations in Madden–Julian oscillation (Wheeler and Hendon 2004). If we are concerned with the PNAI during *n* years, the monthly PNAI in month *j* (*j* is one of the 5 months of NDJFM) of year *i* is calculated by:

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

where $Z^{*\prime}_{i,j} = \frac{Z'_{i,j}}{\sqrt{\frac{1}{n\times 5}\sum_{i=1}^{n}\sum_{j=1}^{5} {Z'_{i,j}}^2}}$ 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 $Z^{*\prime}_{i,j}$ denotes the standardized anomaly of geopotential height at 700 hPa in month *j* of year *i* with seasonal-cycle removed.

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

273 positive PNAI values, the negative PNA months (PNA–) that are 25% of the $n\times5$ PNAI
274 months with the highest negative PNAI values, and the rest months that are referred
275 to as the transitional months (Fig. 2).
276
277 **3 Impacts of PNA on observed aerosol concentrations**
278 The measurements of $PM_{2.5}$ are available over 1999–2013, in which there were 18
279 PNA+ months and 18 PNA– months as shown in Fig. 2a. Figure 3 shows the
280 differences in observed surface-layer $PM_{2.5}$ concentrations between the PNA+ and
281 PNA– months (concentrations averaged over the 18 PNA+ months minus those
282 averaged over the 18 PNA– months). The uncertainty associated with the differences
283 in aerosol concentrations between PNA+ and PNA– months is represented by the
284 two-tail Student-t test with significance level of 90%. Among 1044 sites with $PM_{2.5}$
285 concentrations (Fig.1), 42% of which had statistically significant differences in $PM_{2.5}$
286 between PNA+ and PNA– months. Relative to the PNA– months, $PM_{2.5}$
287 concentrations in PNA+ months were higher in California, the contiguous Salt Lake
288 (northern Utah), and over and near the eastern Midwest. The enhancement of $PM_{2.5}$
289 reached 7–9 μg m^{-3} (or 40–80%) in California, 7–9 μg m^{-3} (80–100%) around the Salt
290 Lake, 3–5 μg m^{-3} (40–80%) over and near the eastern Midwest. At sites in North
291 Dakota, Wisconsin, Michigan, Minnesota, Montana, Texas and Maine, the $PM_{2.5}$
292 concentrations were lower by up to 2 μg m^{-3} (–10 to –20%) in PNA+ months than in
293 PNA– months. As the concentrations are averaged over all sites (including the sites
294 that pass and do not pass the t-test with 90% confidence level) in the U.S., the
295 western U.S. (west of 100°W, Fig.1), and the eastern U.S. (east of 100°W, Fig. 1),
296 $PM_{2.5}$ concentrations were higher by 1.0 μg m^{-3} (8.7%), 1.3 μg m^{-3} (14.3%), and 0.8
297 μg m^{-3} (7.2%), respectively, in the PNA+ months than in PNA– months (Table 1).

298 The measurements of speciated aerosols are available during 2000–2013, in
299 which there were 17 PNA+ and 17 PNA– months (Fig. 2b). Figure 3 also shows the
300 differences in observed surface-layer concentrations of individual aerosol species
301 between the PNA+ and PNA– months. The differences in concentrations of SO_4^{2-},
302 NO_3^- and NH_4^+ show statistically significant positive values at most sites. Among the
303 355, 343, and 194 sites with measurements of SO_4^{2-}, NO_3^- and NH_4^+, 30%, 44%, and
304 39% of which pass the two-tail t-test with 90% confidence level, respectively. While
305 the absolute differences in concentrations of SO_4^{2-}, NO_3^- and NH_4^+ between PNA+
306 and PNA– were in the range of 0–1 μg m^{-3} at most sites, the maximum differences
307 reached 1.5–2.5 μg m^{-3} (30–50%) for SO_4^{2-} in Pennsylvania, 1.5–2.5 μg m^{-3} (150–
308 200%) for NO_3^- in Illinois, Indiana and Ohio, and 1.5–2.5 μg m^{-3} (50–70%) for NH_4^+ in
309 Pennsylvania. Averaged over the sites with measurements, the absolute differences
310 in concentrations of SO_4^{2-} and NO_3^- between PNA+ and PNA– months were larger in
311 the eastern U.S. than in the western U.S.. As shown in Table 1, the differences in the
312 averaged concentrations of SO_4^{2-}, NO_3^- and NH_4^+ were, respectively, 0.1 μg m^{-3}
313 (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}
314 (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
315 0.01 μg m^{-3} (0.5%), 0.3 μg m^{-3} (29.1%), and 0.1 μg m^{-3} (11.9%) in the whole of U.S..
316 With regard to carbonaceous aerosols, among the 105 and 104 sites with
317 measurements of OC and BC, 39% and 31% of which pass the two-tail t-test with 90%
318 confidence, respectively. The differences in concentrations of these two species
319 between PNA+ and PNA– months show similar geographical pattern, with positive
320 values at most sites but negative values in Michigan, New York, and the South
321 Atlantic States. The maximum differences between the PNA+ and PNA– months
322 reached 2.5–3 μg m^{-3} (50–70%) in Kentucky for OC. Averaged over sites with

measurements available, the absolute differences in OC and BC concentrations between the PNA+ and PNA– months were larger in the western U.S. than in the eastern U.S. (Table 1). Among all aerosol species listed in Table 1, OC exhibited the largest absolute differences between the PNA phases in the western U.S., because OC accounts for 25–65% of $PM_{2.5}$ in the western U.S. (Malm et al. 2004) and the OC observed by EPA-AQS network, which are located in urban and suburban settings, were higher than the observations by other long term networks in U.S..(Malm et al. 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.

We have also calculated the correlation coefficient between PNAI and EPA-AQS surface aerosol concentrations at each site for each aerosol species ($PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, OC, or BC) (see auxiliary Fig. S2). At most sites, positive (negative) correlation coefficients in Fig. S2 corresponded to the increases (decreases) in aerosol concentration in PNA+ months relative to PNA– months shown in Fig. 3. Positive correlation coefficients 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 quantified approximately by the square of correlation coefficient (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 $PM_{2.5}$, 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.

4 Impacts of PNA on simulated aerosol concentrations

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-}, NO_3^-, NH_4^+, 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 similar to that of $PM_{2.5}$, with higher concentrations in the eastern U.S. (1–8 μg m^{-3}) than in the western U.S. (0–3 μg m^{-3}) due to the coal-fired power plants in the 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 μg m^{-3}, respectively. The maximum OC concentrations were simulated to be 2–3 μg m^{-3} in two regions, from Ohio to Massachusetts and from Alabama to South Carolina. The simulated BC concentrations in the U.S. were 0–1 μg m^{-3}, except for the contiguous New York where BC concentrations reached 1–2 μg m^{-3}. The magnitudes and geographic distributions of SO_4^{2-}, NO_3^-, NH_4^+ concentrations simulated in our work are similar to

372 those simulated by Park et al. (2006) and Pye et al. (2009), and our simulated OC
373 and BC were similar to those reported by Park et al. (2003).
374 Figure 4b presents the scatter plots of the simulated concentrations versus the
375 EPA-AQS observations. The simulated $PM_{2.5}$ concentrations had normalized mean
376 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
377 simulated and observed aerosol concentrations in month m, respectively. M is the
378 total number of winter months examined) of –30% over the U.S., and the correlation
379 coefficient between simulated and observed $PM_{2.5}$ concentrations was 0.57. The
380 simulated wintertime SO_4^{2-}, NO_3^- and NH_4^+ had NMBs of 37%, 4%, and 26%,
381 respectively. Similar bias in simulated SO_4^{2-} in December-January-February (DJF)
382 was reported Park et al. (2006), as the GEOS-Chem model results were compared
383 with observations from the Clean Air Status and Trends Network (CASTNET). The
384 high bias in our simulated NH_4^+ was associated with the overestimation of SO_4^{2-}. Our
385 model underestimates $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, OC and BC in the western U.S. (Fig.
386 4b), which can be explained in part by the relatively high aerosol concentrations
387 observed for this region from the EPA-AQS. Hand et al. (2014) compared the
388 observed concentrations of aerosols from the EPA-AQS with those from the
389 Interagency Monitoring of Protected Visual Environments (IMPROVE) for 2008–2011,
390 and showed that the ratios of wintertime aerosol concentrations of ammonium sulfate,
391 ammonium nitrate, OC, and BC from the EPA-AQS to those from the IMPROVE were,
392 respectively, 2.3, 7.7, 8.3, and 13.1, as the concentrations were averaged over the
393 western U.S.. Liu et al. (2004) also attributed the high EPA-AQS concentrations in the
394 western U.S. to the relative sparse urban sites that were heavily influenced by strong
395 local sources such as automobiles and wood fires. The low model biases in the
396 western U.S. may also be caused by the biases in emissions in the model.

Since this study is dedicated to examine the influence of PNA phase on the month-to-month variations of aerosol concentrations during wintertime, Fig. 4c compares, for each aerosol species, the deviation from the mean (DM) of observed 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 average aerosol concentration over the U.S. in month m, and M is the number of 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). The model captures fairly well the peaks and troughs of DMs for $PM_{2.5}$, SO_4^{2-}, NO_3^- and NH_4^+, with correlation coefficients of 0.61, 0.45, 0.33, and 0.65, respectively. The model does not capture well the monthly variations of DMs of concentrations of OC and BC, because both anthropogenic and biomass burning emissions are fixed at 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). Since the biomass burning emissions, which contribute largely to carbonaceous aerosols, have large interannual variations (Duncan et al., 2003; Generoso et al., 2003; van der Werf et al., 2006), we also show in Fig. 4c the time series of DMs of biomass burning emissions of OC and BC by using biomass burning emissions in NDJFM over 2000–2006 from GFED v2. The correlation coefficients between biomass burning emissions and observed concentrations of OC and BC were 0.36 and 0.34, respectively, indicating that the observed variations in OC and BC were influenced by monthly and interannual variations in biomass burning. We have also calculated the temporal correlation coefficient between EPA-AQS observations and GEOS-Chem model results at each site for each aerosol species (auxiliary Fig. S4). The temporal correlations were statistically significant for $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+ at

most sites in the U.S., especially over and near the eastern Midwest where largest increases in aerosol concentrations were identified in the PNA+ months relative to the PNA– months.

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– 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 that the pattern of simulated differences in $PM_{2.5}$ between PNA+ and PNA–months was similar to that of observations (Fig. 3), except that the simulated differences were not large in California, mainly due to the underestimation of OC in California as compared to EPA-AQS data (Fig. 4b). The simulated $PM_{2.5}$ concentrations were higher by 0.6 μg m^{-3} (12.2%), 0.3 μg m^{-3} (14.0%), and 0.9 μg m^{-3} (10.8%) over the whole of, western, and eastern U.S., respectively, in the PNA+ months than in PNA– months (Table 2). The simulated relative difference in $PM_{2.5}$ was close to that from 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..

447 Fig. 5 also shows the differences in simulated surface-layer concentrations of
448 SO_4^{2-}, NO_3^- and NH_4^+ between the PNA+ and the PNA– months. The differences in
449 concentrations of SO_4^{2-} were larger in the western than in the eastern U.S., with
450 maximum enhancements of 0.4–0.8 μg m^{-3} (30–50%) over the West North Central
451 States (South Dakota, Nebraska, Minnesota, Iowa, and Missouri). The differences in
452 concentrations of NO_3^- and NH_4^+ between PNA+ and PNA– months had similar
453 geographical patterns, with increases in concentrations in a large fraction of the
454 eastern U.S. and over a belt region along the Rocky Mountain in the western U.S..
455 The increases in concentrations of NO_3^- and NH_4^+ over the eastern U.S. in PNA+
456 months relative to PNA– months agreed very well with those seen in observations
457 (Fig. 3). The differences in concentrations of SO_4^{2-}, NO_3^- and NH_4^+ in the eastern U.S.
458 between the PNA+ and PNA– months were 0.2 μg m^{-3} (4.0%), 0.4 μg m^{-3} (33.5%),
459 and 0.2 μg m^{-3} (13.2%), respectively (Table 2).
460 The differences in concentrations of OC and BC between PNA+ and PNA–
461 months had similar geographical patterns, with large increases in concentrations over
462 and near the eastern Midwest and the region from northwestern U.S. to Texas. The
463 maximum differences reached 0.2–0.4 μg m^{-3} (10–20%) and 0.1–0.2 μg m^{-3} (20–
464 30%) in Illinois, Indiana and Ohio for OC and BC, respectively. The magnitudes of the
465 differences in OC and BC were statistically significant but were smaller than the
466 observations (Tables 1 and 2). The absolute differences in OC were less than 0.1 μg
467 m^{-3} in the western, eastern and whole of U.S due to the underestimation of OC in the
468 simulation.
469 In summary, model results agreed with observations in that the concentrations of
470 all aerosol species of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC averaged over the U.S. were
471 higher in PNA+ months than in PNA– months. Relative to the PNA– months, the

472 average concentration of $PM_{2.5}$ over the U.S. was higher by about 8.7% (12.2%)
473 based on observed (simulated) concentrations. Furthermore, simulated geographical
474 patterns of the differences in $PM_{2.5}$ and each aerosol species between the PNA+ and
475 PNA– months were similar to those seen in observations, with the largest increases in
476 aerosol concentrations in PNA+ months over and near the eastern Midwest.
477
478 **5 Mechanisms for the impact of PNA on aerosol concentrations**
479 **5.1 The impact of PNA on transboundary transport of aerosols**
480 The transboundary transport of pollutants to and from the U.S. depends largely on
481 winds in the free troposphere (Liang et al. 2004). Figure 6 shows the horizontal winds
482 at 700 hPa averaged over the winter months of NDJFM of 1986–2006 and the
483 corresponding differences between PNA+ and PNA– months on the basis of the
484 GEOS-4 meteorological fields. Strong westerlies prevailed over the U.S. in wintertime
485 (Fig. 6a). Relative to the PNA– months, anomalous northeasterlies occurred over a
486 large fraction of U.S. in the PNA+ months. Anomalous anti-cyclonic circulation
487 occurred near the northwestern U.S. and anomalous cyclonic circulation occurred
488 near the southeastern U.S. (Fig. 6b), corresponding to the large positive and negative
489 differences in geopotential height in these two regions (see auxiliary Fig. S1),
490 respectively.
491 We also calculate mass fluxes of $PM_{2.5}$ at the four lateral boundaries (from the
492 surface to 100 hPa) of the U. S. for different PNA phases (Table 3). The domain of the
493 box to represent the U. S. is (75–120°W, 28–49°N), as shown in Fig. 6b. For $PM_{2.5}$ in
494 wintertime, the inflow from the west boundary and the outflow from the east boundary
495 had the largest absolute values (Table 3). Relative to the PNA– months, the inflow
496 from the west boundary and the outflow from the east boundary in PNA+ months

497 exhibited reductions of 17.1 kg s^{-1} and 12.8 kg s^{-1}, respectively (Table 3). The inflow
498 flux from south boundary decreased by 19.3 kg s^{-1}, and the inflow flux from north
499 boundary increased by 37.6 kg s^{-1}, leading to a net increase of inflow flux of 14.0 kg
500 s^{-1} in PNA+ months. Therefore, the transboundary transport has an overall effect of
501 increasing $PM_{2.5}$ aerosols in the U.S. in PNA+ months relative to PNA– months. The
502 relative change in net flux was 11.4% ((PNA+ minus PNA–)×100%/PNA–) (Table 3),
503 coinciding well with the enhancement of 12.2% in surface-layer $PM_{2.5}$ concentration
504 averaged over the U.S. (Table 2). Note that because the GEOS-Chem model
505 underestimates $PM_{2.5}$ concentrations in the western U.S. (Fig. 4b), the net outflow
506 flux from the selected box might have been underestimated, but this should not
507 compromise our conclusions about the differences in net flux between PNA+ and
508 PNA– phases.

509 **5.2 Local changes in aerosol concentrations caused by the PNA**

510 The PNA pattern is also associated with variations in meteorological variables such
511 as temperature (Leathers et al. 1991; Konrad II 1998; Notaro et al. 2006; Knight et al.
512 2008; Liu et al. 2015; Ning and Bradley 2014, 2015), precipitation (Leathers et al.
513 1991; Henderson and Robinson 1994; Coleman and Rogers 2003; Notaro et al. 2006;
514 Archambault et al. 2008; Myoung and Deng 2009; Ning and Bradley 2014, 2015;
515 Wise et al. 2015) and humidity (Sheridan 2003; Coleman and Rogers 2003; Knight et
516 al. 2008) in U.S.，which are expected to influence aerosol concentrations within the
517 U.S. through chemical reactions, transport, and deposition.

518 Figure 7 shows the composite differences, between the PNA+ and PNA– months,
519 in surface air temperature (T), precipitation rate (PR), relative humidity (RH), surface
520 wind speed (WS), and planetary boundary layer height (PBLH), based on the
521 reanalyzed GEOS-4 datasets. Relative to PNA– months, temperatures were higher

522 by 1–3 K over the northwestern U.S. and lower by 1–4 K in the southeastern region.
523 Such geographic distributions of temperature anomalies were attributed to the
524 maritime warm air in the northwestern U.S. accompanied by the enhanced
525 tropospheric geopotential height in North America (Leathers et al. 1991; Sheridan
526 2003; Liu et al. 2015; Ning and Bradley 2015) (see also auxiliary Fig. 1d) and the
527 more frequent outbreak of cold air in southeastern U.S. accompanied by the
528 depressed geopotential height (Konrad II 1998; Liu et al. 2015) (see also auxiliary Fig.
529 1d). The differences in precipitation between PNA+ and PNA– months reached –1.6
530 to –2.4 mm day^{-1} (–32 to –48%) over and near the eastern Midwest, –2.4 to –3.2 mm
531 day^{-1} (–48 to –64%) in the northwestern U.S., and 1.6–2.4 mm day^{-1} (16–32%) in the
532 southeastern U.S.. These effects of PNA on precipitation were similar to those
533 obtained from wintertime station data by Leathers et al. (1991), Coleman and Rogers
534 (2003) and Wise et al. (2015). With respect to RH, the values in the eastern U.S. were
535 generally lower in the PNA+ months than in PNA– months, as a result of the reduced
536 moisture flux from the Gulf of Mexico to the eastern U.S. (Coleman and Rogers 2003),
537 where RH showed maximum reduction of –3 to –9%. The enhancement of RH of up
538 to 6–9% in Texas, Oklahoma and New Mexico was due to the anomalous easterlies
539 over the south central U.S. (see Fig. 7), which diminished the influence of the dry air
540 from the deserts of the southwestern U.S. and northwestern Mexico in PNA+ months
541 (Sheridan 2003). The surface WS showed reductions in PNA+ months relative to
542 PNA– months in two regions, one was the belt region along the Rocky Mountain from
543 the northwestern U.S. to Texas (with the maximum reductions of 1.5–2.0 m s^{-1} (48–
544 64%)) and the other region, with a northeast-southwest orientation, was from Ohio to
545 Louisiana (with maximum reductions of 0.5–1.0 m s^{-1} (16–32%)). The differences in
546 PBLH between PNA+ and PNA– months were statistically significant in the western

547 U.S. and in the belt region from the northeastern U.S. to the eastern Midwest, with the
548 maximum reductions in PBLH of 75–100 m (15–20%) and 75–100 m (10–15%),
549 respectively.

550 The comparisons of Fig. 5b with Fig. 7a indicate that the increases in $PM_{2.5}$
551 concentrations over and near the eastern Midwest in PNA+ months relative to the
552 PNA– months can be attributed to the decreases in PR, WS and PBLH in these
553 locations, since the changes in these three variables depressed wet deposition, local
554 horizontal diffusion, and vertical diffusion of the surface aerosols, respectively. The
555 increases in SO_4^{2-} in the western U.S. (Fig. 5b) corresponded to the decreases in PR
556 and PBLH and the increased temperature. The increases in temperature enhance
557 chemical reaction rates (Aw and Kleeman 2003; Dawson et al. 2007; Kleeman 2008).
558 In the eastern U.S., although PR, WS, and PBLH decreased over and near the
559 eastern Midwest, the cooling in the eastern U.S. might have offset the effects by PR,
560 WS, and PBLH, inducing practically no changes in SO_4^{2-} in the eastern U.S.. The
561 large increases in NO_3^- and NH_4^+ in the southeastern U.S. (Fig. 5b) can be attributed
562 to the reduced surface temperature, which was favorable to the wintertime formation
563 of NO_3^- and NH_4^+ (Dawson et al. 2007). The differences in concentrations of OC and
564 BC between PNA+ and PNA– corresponded well with the reduced PR, WS and
565 PBLH.

566 In order to quantify the impacts of anomalies in meteorological parameters driven
567 by PNA on concentrations of different aerosol species, the pattern correlation
568 coefficients (PCC, http://glossary.ametsoc.org/wiki/Pattern_correlation) are
569 calculated and shown in Table 4. These pattern correlation coefficients denote the
570 relationship between the geographical distribution of anomalies of each of T, PR, RH,
571 WS, and PBLH (Fig. 7b) and that of the differences in concentration of each aerosol

572 species between PNA+ and PNA– months (Fig. 5c). As shown in Table 4, over the
573 whole U.S., the PNA influenced SO_4^{2-} concentrations mainly through changes PBLH,
574 PR, and T, with the highest PCC values of –0.43, –0.38, and +0.26, respectively. For
575 NO_3^-, the PNA-induced variations in temperature had a strong negative correlation
576 (PCC=–0.59) with the PNA-induced differences in concentrations, indicating that
577 surface temperature was the dominant meteorological factor to influence NO_3^-
578 concentrations. For NH_4^+, OC, and BC, PR and PBLH were the two variables that had
579 the largest negative PCC values (Table 4). Note that PBLH was the most important
580 meteorological variable for SO_4^{2-}, NH_4^+, OC and BC, which contributed to the high
581 correlation between PBLH and $PM_{2.5}$.
582
583 **6 Conclusions**
584 This study examined the impacts of monthly variation in the PNA phase on wintertime
585 surface-layer aerosol concentrations in the U.S. by the analyses of EPA-AQS
586 observations over 1999–2013 and model results for 1986–2006 from the global
587 chemical transport model GEOS-Chem.
588 The composite analyses on the EPA-AQS observations showed that the average
589 concentrations of $PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, OC, and BC aerosols over the U.S. were
590 higher in the PNA+ months than in the PNA– months by 1.0 μg m^{-3} (8.7%), 0.01 μg
591 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
592 m^{-3} (27.8%), respectively. Regionally, the observed $PM_{2.5}$ concentrations were higher
593 by 3–5 μg m^{-3} (40–80%) over the Midwest, and by 7–9 μg m^{-3} (80–100%) around the
594 Salt Lake, as the concentrations in PNA+ months were compared to those in PNA–
595 months.

596 The impacts of PNA phase on aerosol concentrations were reproduced fairly well
597 by the GEOS-Chem simulation with fixed anthropogenic emissions (the variations of
598 aerosols concentrations were driven by changes in meteorological fields alone).
599 Concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC averaged over the U.S. were
600 simulated to be higher in the PNA+ months than in PNA– months. The average
601 concentration of $PM_{2.5}$ over the U.S. was simulated to be 12.2% higher in the PNA+
602 months relative to the PNA– months, in close agreement with the observations.
603 Simulated geographical patterns of the differences in $PM_{2.5}$ and each aerosol species
604 between the PNA+ and PNA– months were similar to those seen in observations. The
605 largest increases in aerosol concentrations in the PNA+ months were simulated to be
606 over and near the eastern Midwest, but the model results showed small PNA-induced
607 changes in aerosol concentrations in the western U.S..
608 The mechanisms for the impacts of PNA on aerosol concentrations were
609 examined. The transboundary transport was found to have an overall effect of
610 increasing $PM_{2.5}$ aerosols in the U.S. in the PNA+ months relative to PNA– months.
611 Compared to the PNA– months, anomalous northeasterlies occurred over a large
612 fraction of U.S., which led to a net increase in inflow flux of $PM_{2.5}$ of 14.0 Kg s^{-1} in
613 PNA+ months. Regionally within the U.S., the PNA influenced aerosol concentrations
614 through changes in precipitation rate (PR), planetary boundary layer height (PBLH),
615 surface wind speed (WS), surface air temperature (T) and relative humidity (RH), as
616 represented by the pattern correlation coefficients (PCCs). The PNA influenced SO_4^{2-}
617 concentration mainly through changes PBLH, PR, and T, with the highest PCC values
618 of –0.43, –0.38, and +0.26, respectively. For NO_3^-, the PNA-induced variations in
619 temperature had a strong negative correlation (PCC=–0.59) with the PNA-induced
620 differences in concentrations. For NH_4^+, OC, and BC, PR and PBLH were the two

621 variables that had the largest negative PCC values. Because PBLH was the most
622 important meteorological variable for SO_4^{2-}, NH_4^+, OC and BC, which led to a high
623 correlation between PBLH and $PM_{2.5}$.
624 Conclusions from this study have important implications for air quality in the U.S..
625 Leathers and Palechi (1992) showed that the PNAI were generally low in 1947–1957
626 but consistently high in 1958–1987. The PNAI during 1948–2010 exhibited an
627 increasing trend for positive phases and a decreasing trend for negative phases (Liu
628 et al. 2015; Ning et al. 2015; http://research.jisao.washington.edu/data_sets/pna/#djf),
629 indicating that wintertime particulate matter pollution in most areas of U.S.
630 deteriorated due to variations in PNA phase alone. Climate models projected that
631 positive PNA phases would increase in the future because of the global warming
632 (Kachi and Nitta, 1997; Müller and Roeckner 2008; Zhou 2014). Therefore, the trend
633 in PNA pattern underlies the necessity of strict emission reduction strategies for
634 greenhouse gases, aerosols, and aerosol precursors.
635
636
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Table 1. The absolute (μg 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 (μg 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 100 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
PNA+	West	89.7
	East	259.3
	South	24.4
	North	–36.2
	Net flux	–109.0
PNA–	West	106.8
	East	272.1
	South	43.7
	North	1.4
	Net flux	–123.0
Diff. (PNA+ minus PNA–)	West	–17.1
	East	–12.8
	South	–19.3
	North	–37.6
	Net flux	14.0

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
T	–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*

Figure Captions

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).

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) 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 calculated using the GEOS-4 data (GEOS4-PNAI), with the PNA+ and PNA– months over 1986–2006 indicated. Red circles are PNA+ months and blue circles PNA– months.

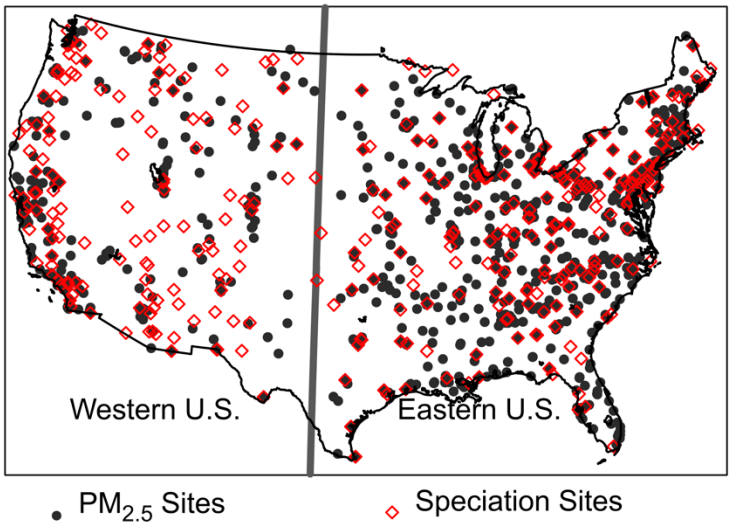
Fig. 3. The absolute (μg m^{-3}, left column) and relative differences (%, right column) in observed monthly mean aerosol concentrations between PNA+ and PNA– months (PNA+ minus PNA–). The measurements of $PM_{2.5}$ were carried out over 1999–2013, in which there were 18 PNA+ months and 18 PNA– months as shown in Fig. 2a. The measurements of speciated aerosols were taken during 2000–2013, in which there were 17 PNA+ and 17 PNA– months (Fig. 2b). The sites with black dots were the differences that passed the two-tail t-test with 90% confidence level.

Fig. 4. (a) Simulated surface-layer concentrations (μg m^{-3}) of $PM_{2.5}$ (the sum of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC) and each aerosol species averaged over NDJFM of 1999–2006. (b) Scatter plots of the simulated concentrations (μg m^{-3}, vertical axis) versus the EPA-AQS observations (μg m^{-3}, horizontal axis). Also shown are the y=x line (black dash line), linear fit for whole U.S. (black line), linear fit for western U.S. (blue line), and linear fit for eastern U.S. (red line). The blue and red dots represent sites in the western and eastern U.S., respectively. (c) Comparisons of the deviation from the mean (DM) of observed concentration (black line) with that of simulated concentration (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.

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.

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.

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.



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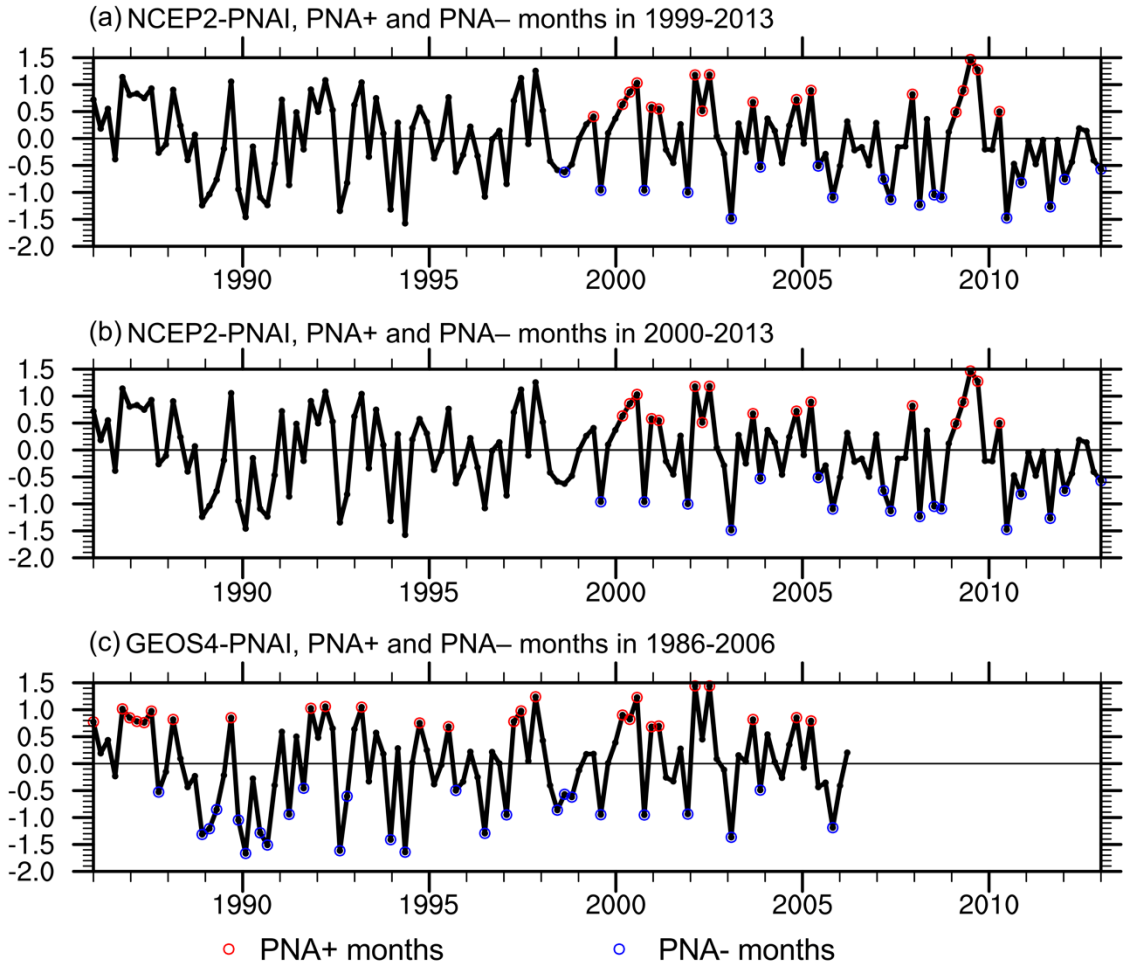

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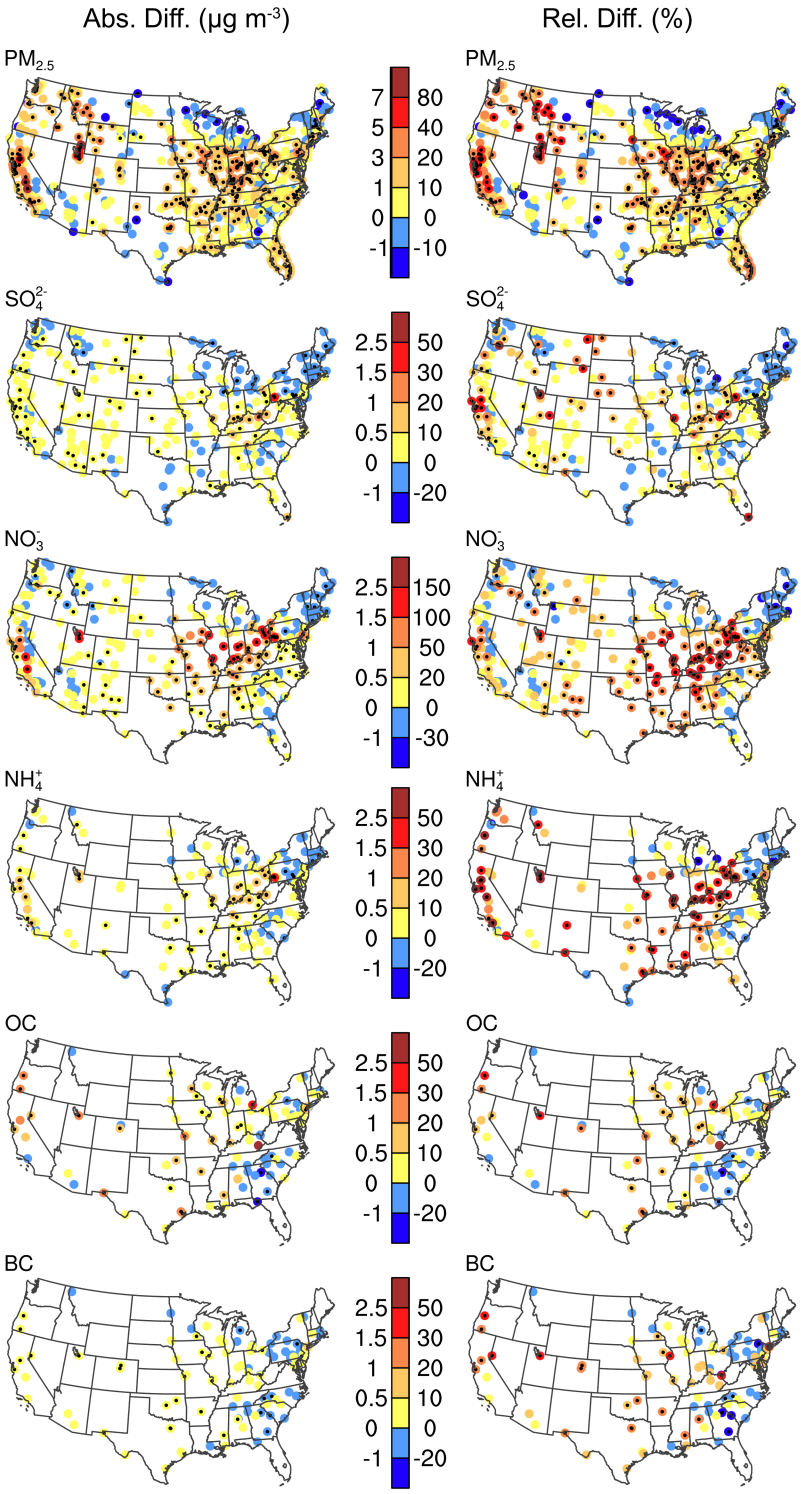

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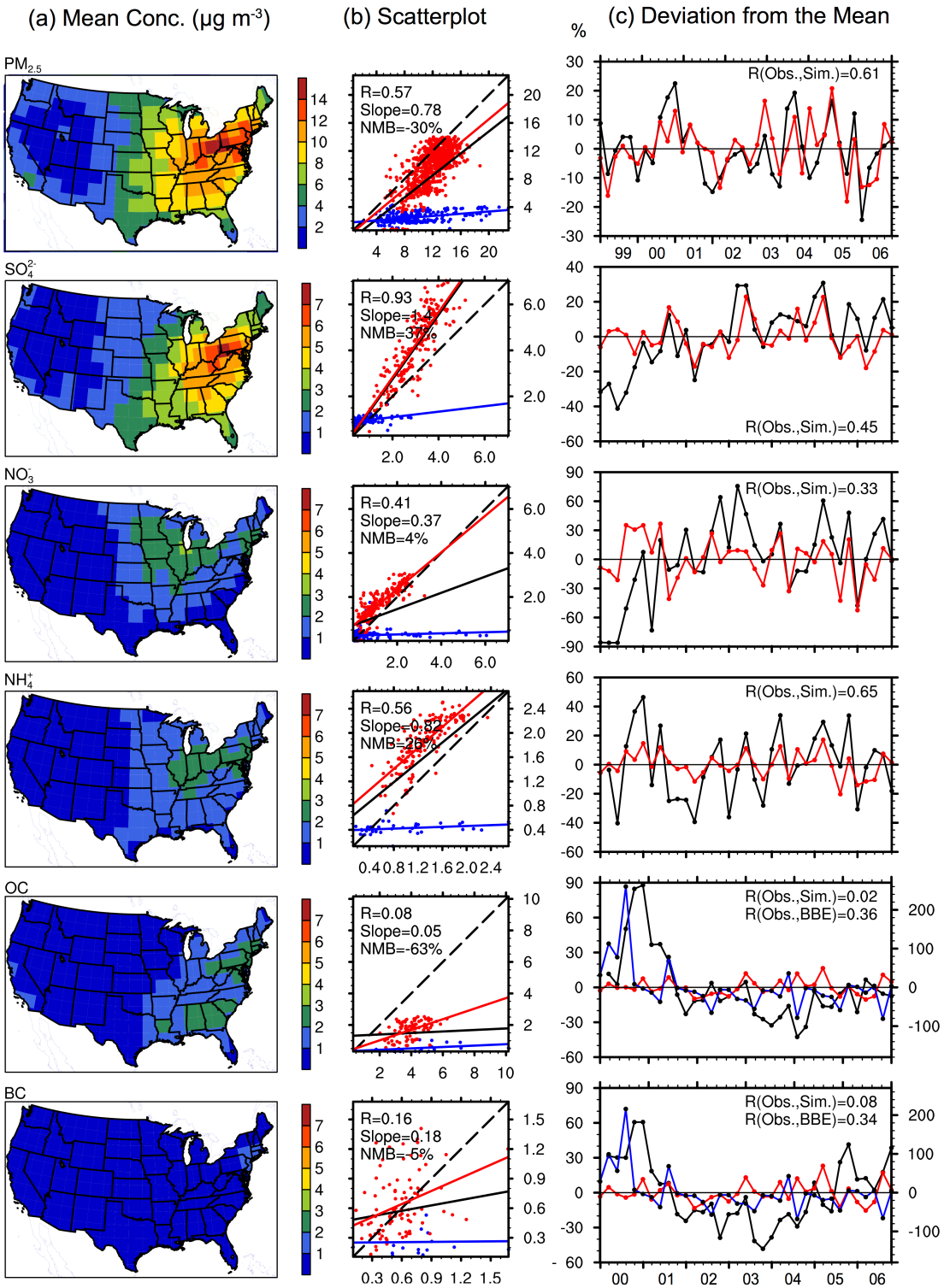

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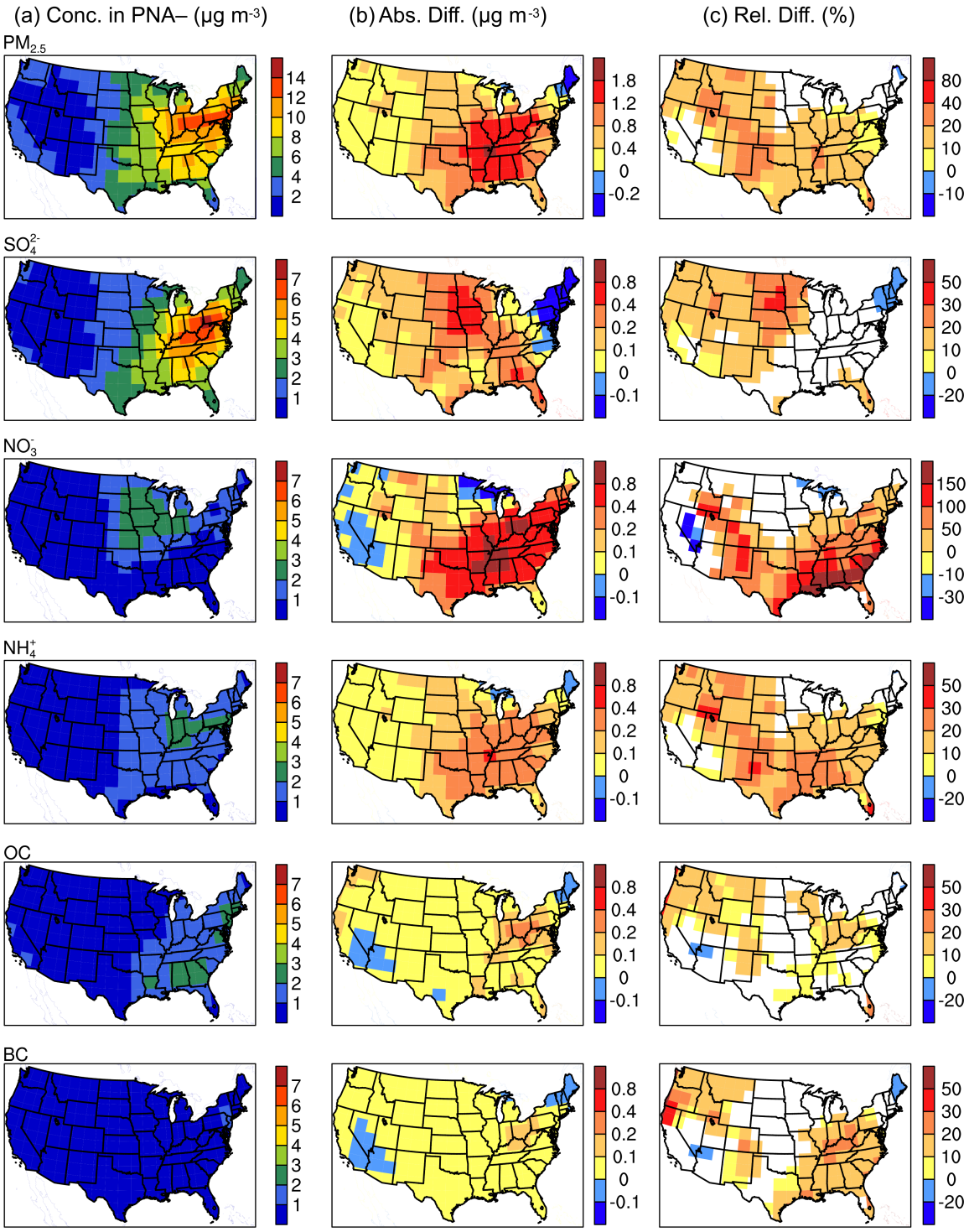

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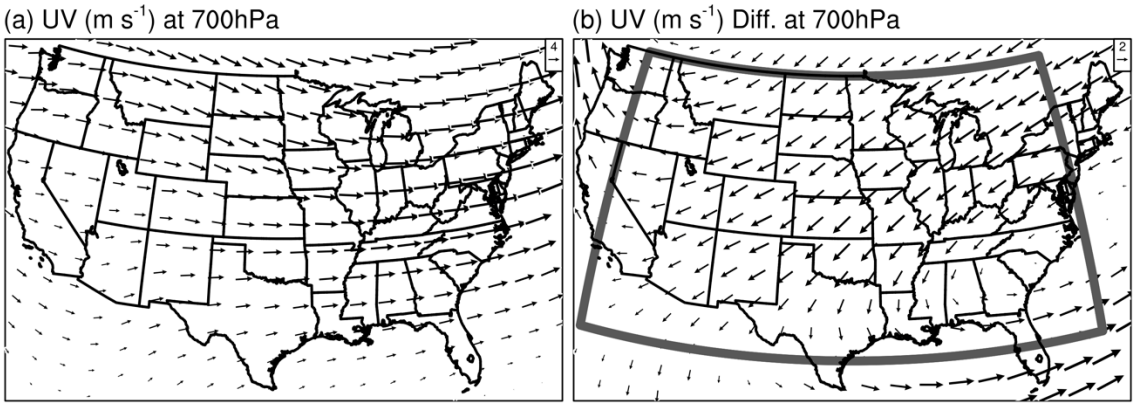

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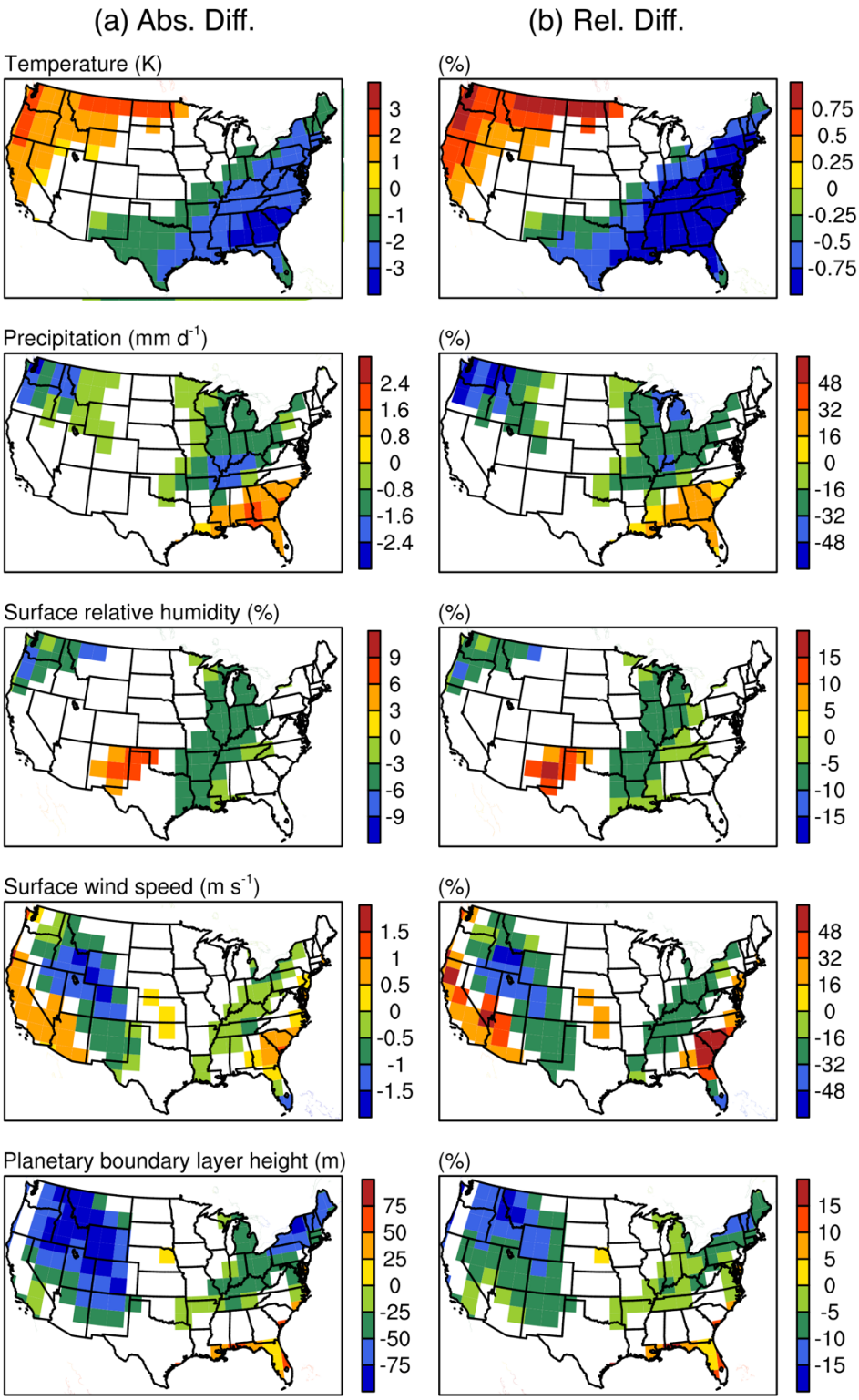

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