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.

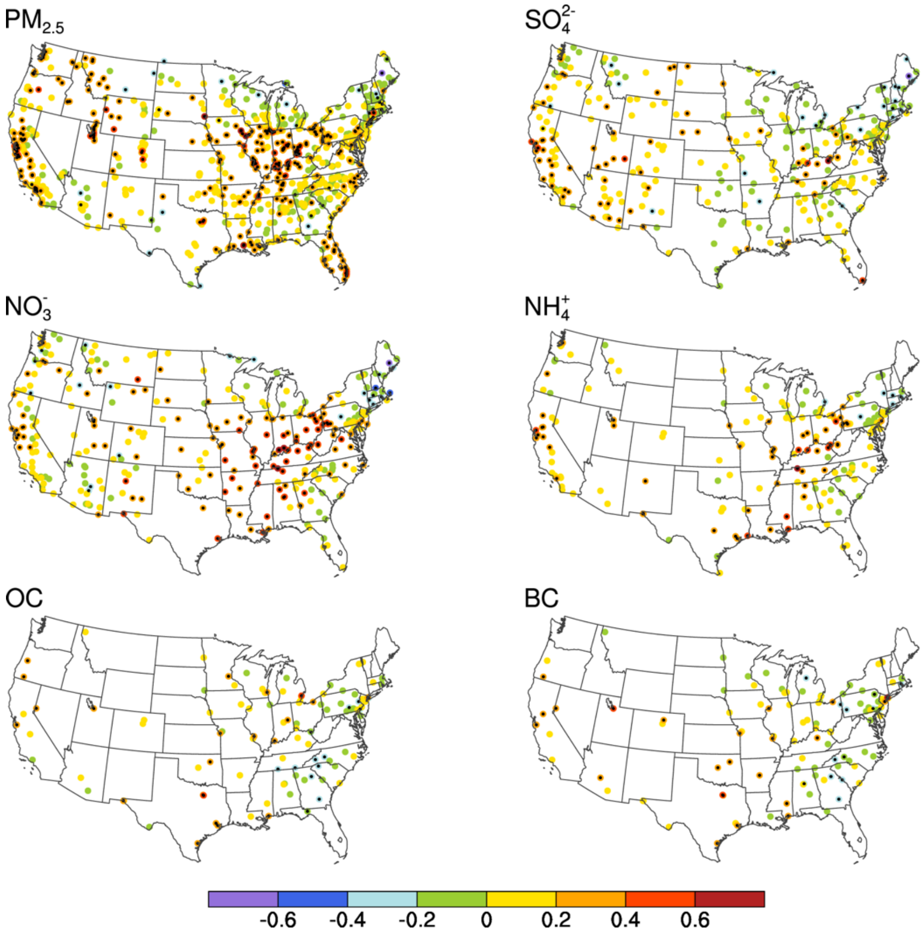

Fig. S2. Correlation coefficient between PNAI and EPA-AQS surface aerosol concentrations at each site for each aerosol species (PM$_{2.5}$, SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, BC, or OC). The measurements are the same as those used in Fig. 3. The sites with black dots are those that have passed the two-tail t-test with 90% confidence level.

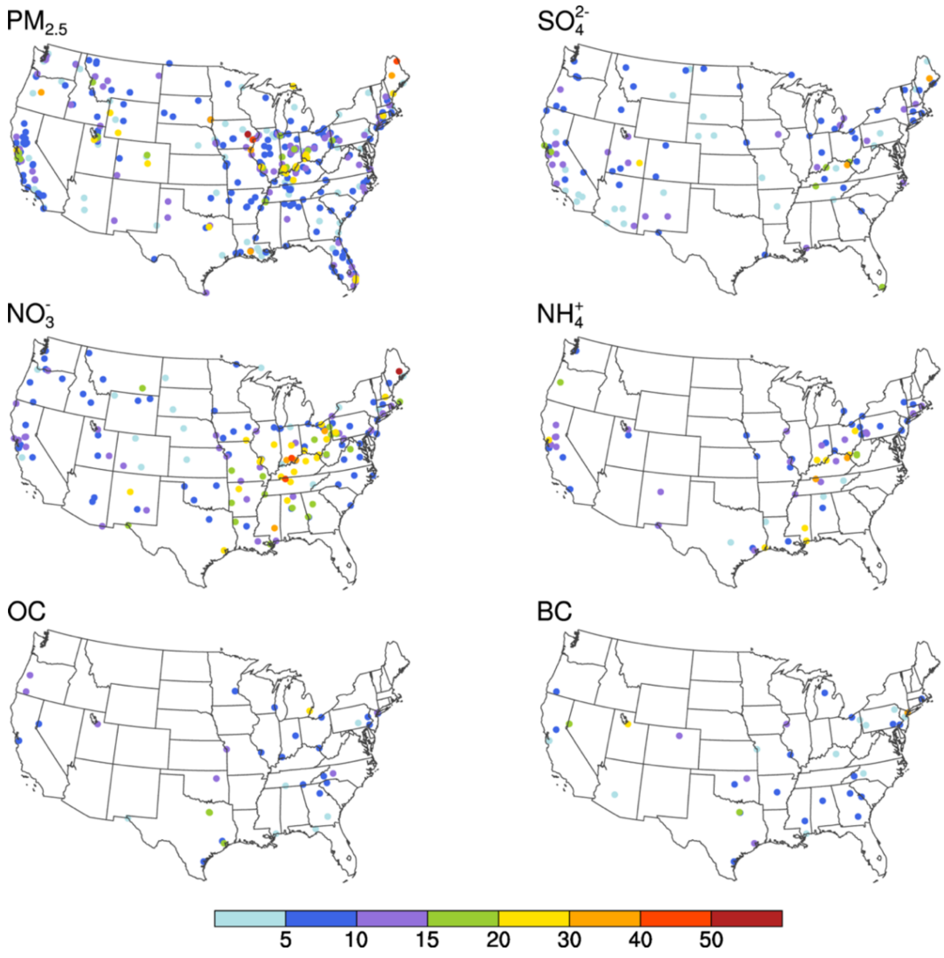

Fig. S3. The fraction (%) of temporal variability of EPA-AQS surface aerosol concentrations explained by PNA at each site for each aerosol species ($PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, BC, or OC). The measurements are the same as those used in Fig. 3. The sites shown here are those in Fig. S2 with 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) ($\mathrm{PNAI_{Leathers}}$). This is the definition used in our manuscript. (2) PNAI defined by Wallace and Gutzler (1981) ($\mathrm{PNAI_{Wallace}}$) as:

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

, where * denotes the normalized geopotential height at 500 hPa. (3) PANI defined by NOAA Climate Prediction Center (CPC) ($\mathrm{PNAI_{NOAA}}$, http://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/month_pna_index2.shtml). All these three definitions reflect the same atmospheric teleconnections over the region of North Pacific to North America region. The correlation coefficients between $\mathrm{PNAI_{Leathers}}$ and $\mathrm{PNAI_{Wallace}}$ (or between $\mathrm{PNAI_{Leathers}}$ and $\mathrm{PNAI_{NOAA}}$) 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 $\mathrm{PNAI}_{\mathrm{Wallace}}$ (Fig. B below) and $\mathrm{PNAI}_{\mathrm{NOAA}}$ (Fig. C below) are similar to those on the basis of $\mathrm{PNAI}_{\mathrm{Leathers}}$ (Fig. 3 in our manuscript). In Fig. B, the enhancement of $\mathrm{PM}_{2.5}$ 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 $\mathrm{PM}_{2.5}$ 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.

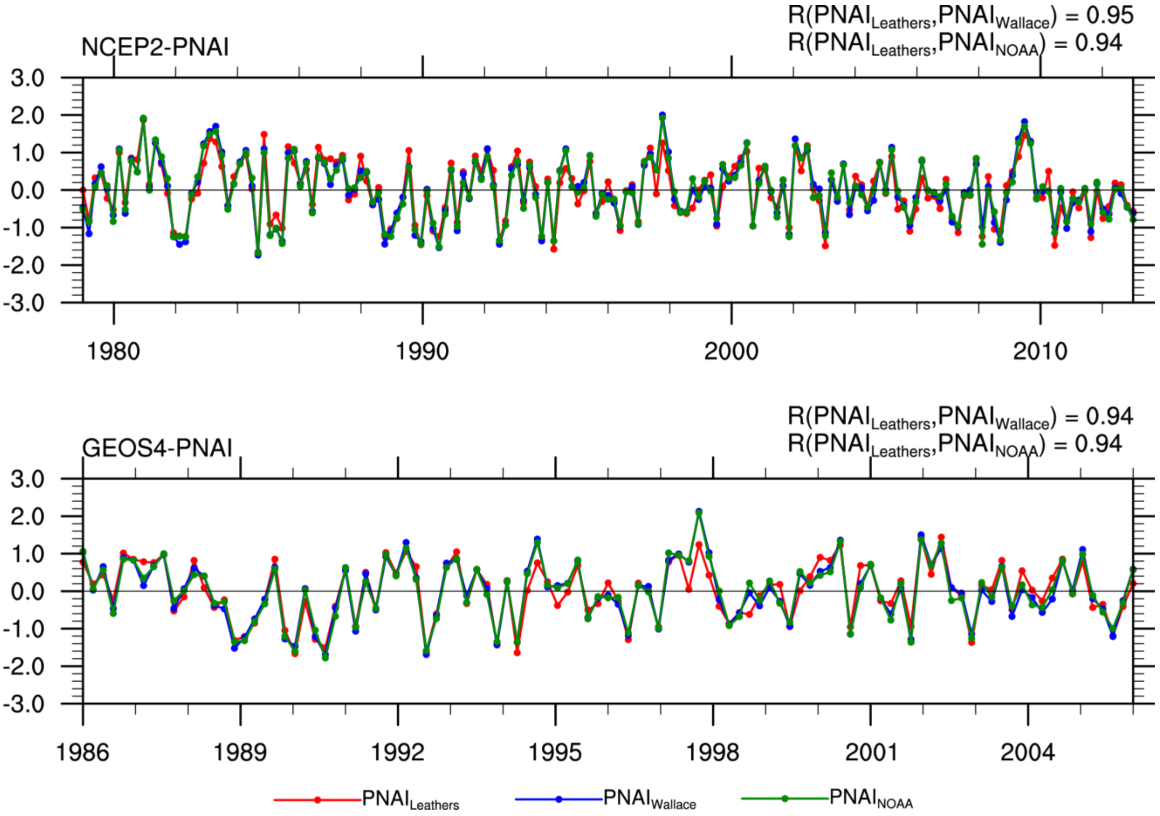

Fig. A. Top panel: Monthly PNAI in NDJFM for years of 1979–2013 calculated using the NCEP-2 data. Bottom panel: PNAI for 1986–2006 calculated using the assimilated GEOS-4 data. The correlation coefficients are marked on top right corner of each panel.

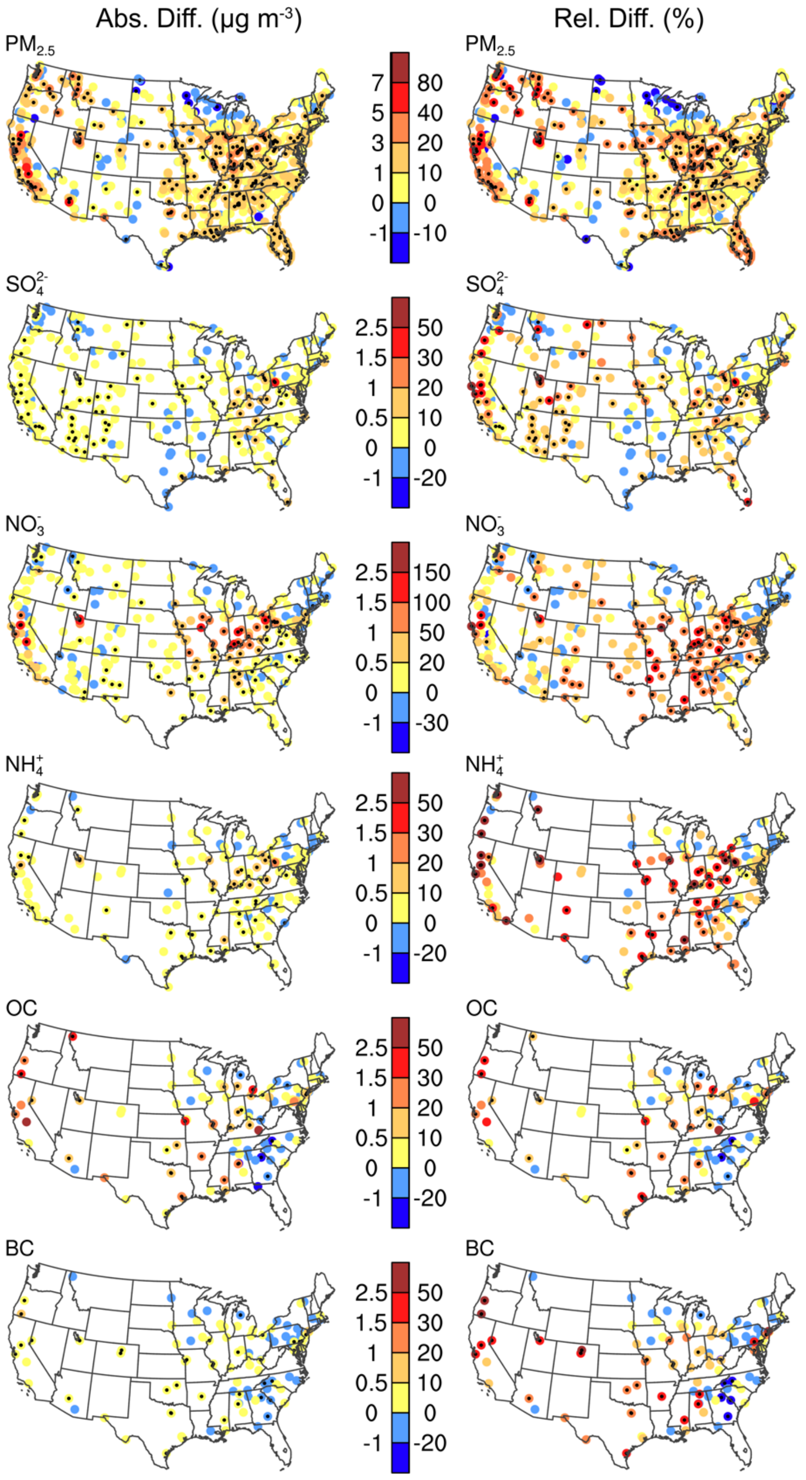

Fig. B. The same as Fig. 3 in the manuscript except that the differences in observed surface-layer aerosol concentrations between the PNA+ and PNA– months are obtained on the basis of PNAI$_{\mathrm{Wallace}}$.

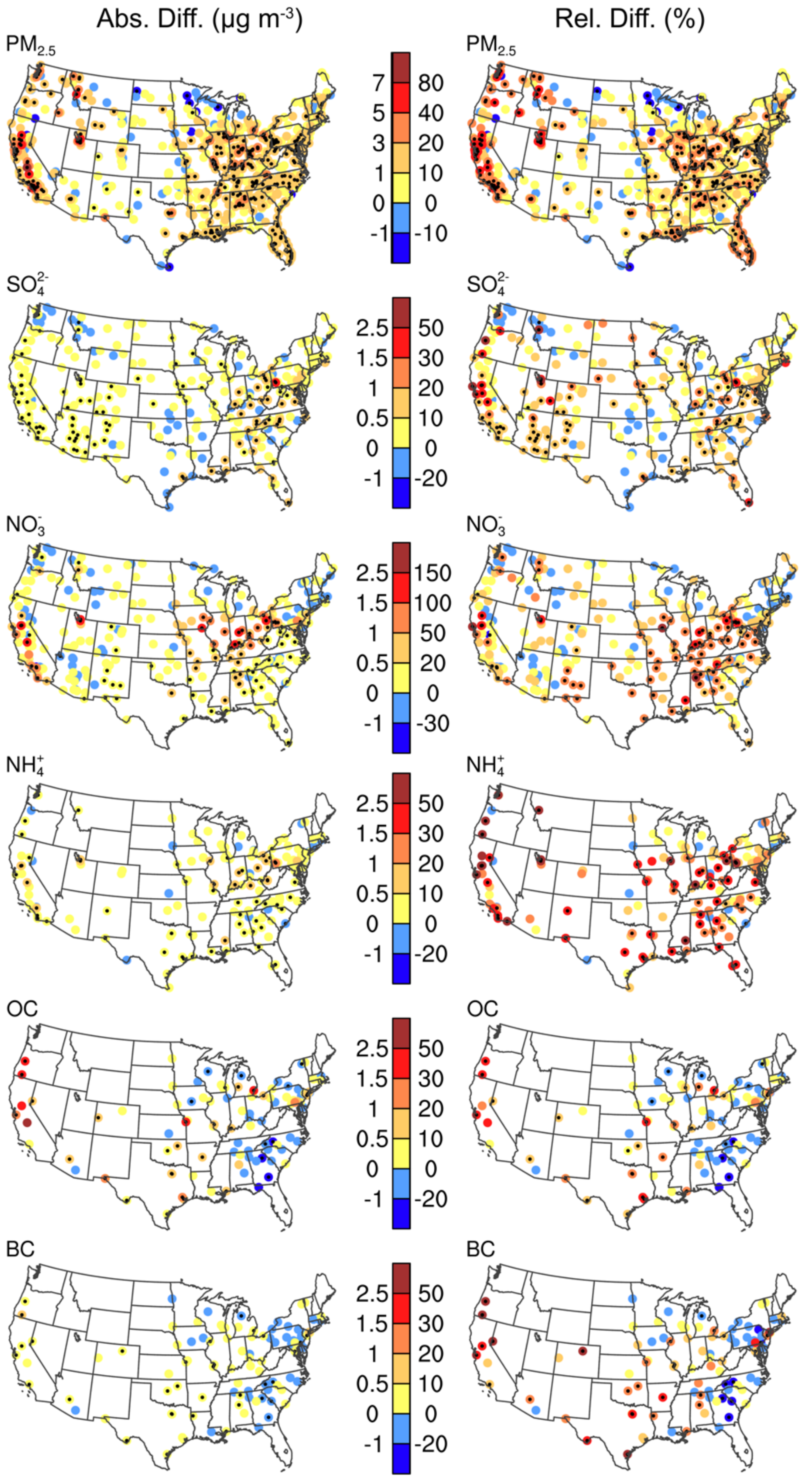

Fig. C. The same as Fig. 3 in the manuscript except that the differences in observed surface-layer aerosol concentrations between the PNA+ and PNA– months are obtained on the basis of $\mathrm{PNAI}_{\mathrm{NOAA}}$.

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 $\mu g\ m^{-3}$) 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.

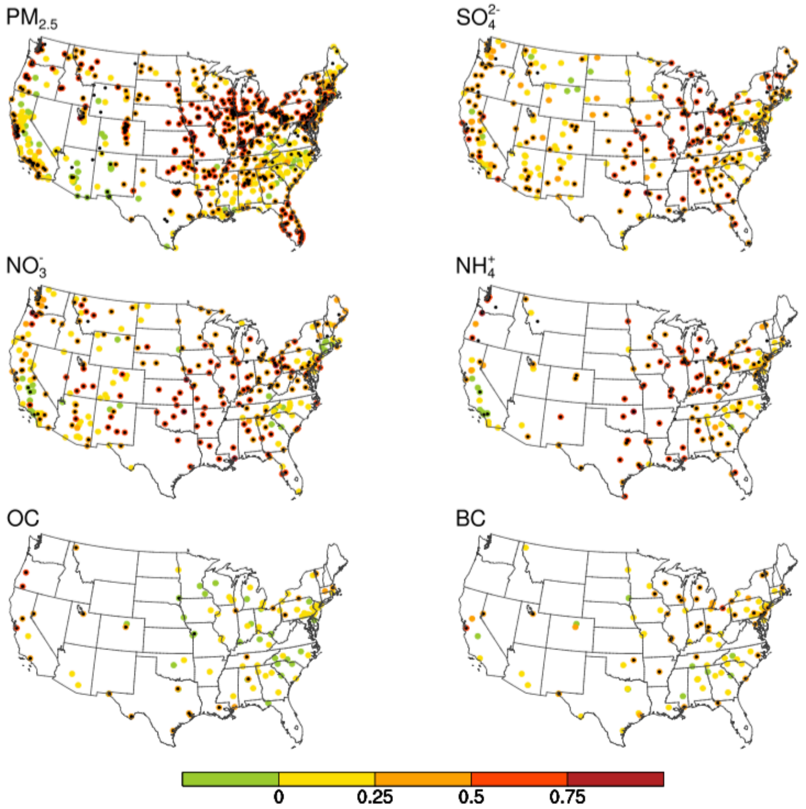

Fig. S4. The temporal correlation coefficient between EPA-AQS observations and GEOS-Chem model results at each site for each aerosol species. Datasets used here are the same as those used in Figure 4 of the manuscript. The sites with black dots are those that have passed the two-tail t-test with 90%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

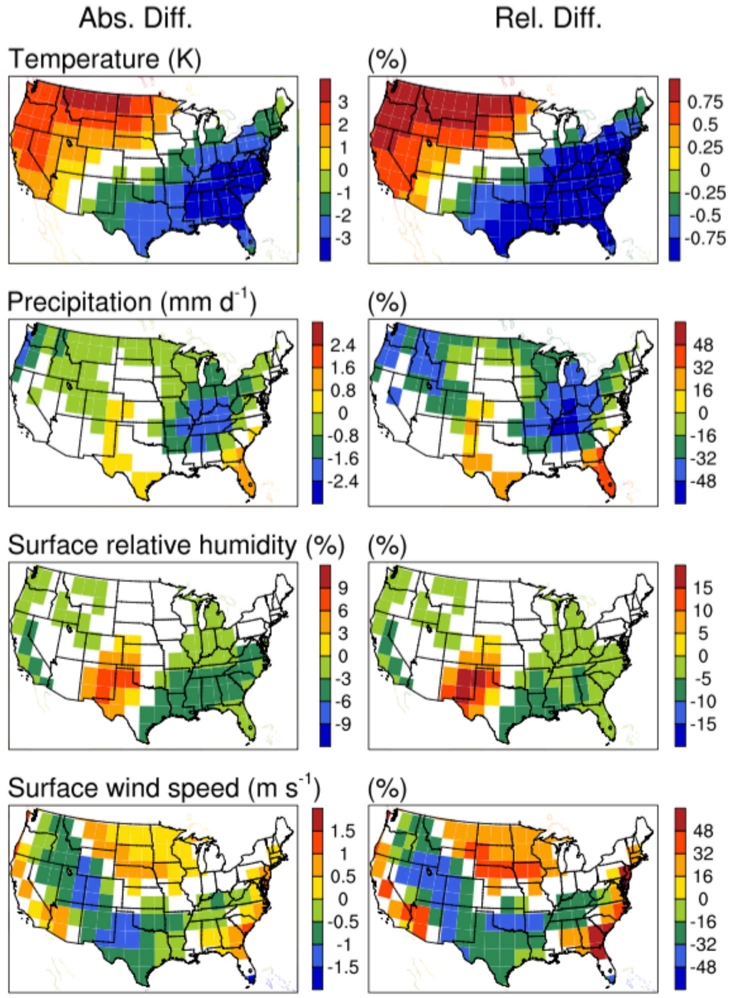

Fig. D. The same as Fig. 7 in the manuscript except that the differences in T, RH, and surface wind speed between PNA+ and PNA– months are calculated for 1948–2014 by using the NCEP Reanalysis and the differences in precipitation are calculated for 1948–2010 by using the Global Precipitation Climatology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

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