

## 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 PM<sub>2.5</sub> 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 CH<sub>4</sub> 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  $11.2 \pm 1.3$  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  $1.18 \times 10^6$  molec  $\text{cm}^{-3}$  ([http://wiki.seas.harvard.edu/geos-chem/index.php/Mean\\_OH\\_concentration](http://wiki.seas.harvard.edu/geos-chem/index.php/Mean_OH_concentration)), which is less than the OH concentration ( $1.22 \times 10^6$  molec  $\text{cm}^{-3}$ ) from the version 9-01-02 by Holmes et al. (2013) and the OH concentration ( $1.24 \times 10^6$  molec  $\text{cm}^{-3}$ ) 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 be 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 ( $\mu g m^{-3}$ ) and relative (%) differences in observed aerosol concentrations between the PNA+ and PNA– months (PNA+ minus PNA–). The observed concentrations are averaged over 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 <sub>2.5</sub>	1.9 (17.6%)**	2.7 (30.5%)**	1.7 (14.9%)**
SO <sub>4</sub> <sup>2-</sup>	0.1 (6.3%)	0.2 (20.3%)**	0.2 (5.4%)
NO <sub>3</sub> <sup>-</sup>	0.6 (68.8%)**	0.5 (67.2%)**	0.7 (71.6%)**
NH <sub>4</sub> <sup>+</sup>	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 PM<sub>2.5</sub>, SO<sub>4</sub>, NO<sub>3</sub>, 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 be 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  $n \times 5$  PNAI months with the highest positive PNAI values, the negative PNA months (PNA-) were 25% of the  $n \times 5$  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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