

We thank Dr. Omid Alizadeh-Choobari and the three anonymous referees for their valuable comments and constructive suggestions on the manuscript. Below, we explain how the comments and suggestions are addressed and make note of the revision we made in the manuscript.

Referee #1

General comments:

- *Using various observational datasets, the present study has evaluated the performance of the quasi-global WRF-Chem model in terms of simulating both meteorological fields and aerosol properties over the Pacific region. Code modifications for a quasi-global WRF-Chem simulation were conducted at the Pacific Northwest National Laboratory (PNNL), and the modifications are planned to be incorporated in the future available release of WRF-Chem. The overall conclusion of the present study is that the model well simulated spatial and seasonal variability of both meteorological fields and aerosol properties across the Pacific region. Apart from running the WRF-Chem model on the quasi-global scale, which has already been conducted and its performance evaluated by Zhao et al. (2013b), the present study does not provide any new insights into the concept of transport of aerosols across the Pacific Ocean. Nevertheless, the observed datasets that have been gathered and the conducted numerical simulation have the potential to extend the current knowledge of the scientific community on the meteorological influences on transport of aerosols across the Pacific Ocean in different seasons. My general comment is major revision of the manuscript, both in the review provided in the introduction and in the analysis of the results. More details are provided below.*

We thank Dr. Omid Alizadeh-Choobari for a detail review. As we stated in the manuscript, “Although the quasi-global WRF-Chem simulation described by Zhao et al. (2013b) has been used to provide realistic chemical lateral boundary conditions for multiple regional modeling studies (e.g., Zhao et al., 2014; Fan et al., 2015), its evaluation has not been documented so far.” and “We focus on the simulation over the trans-Pacific transport region as a first step to evaluate the simulation for providing consistent lateral chemical boundaries for nested regional simulations used to investigate the impact of transported aerosols on regional air quality and climate.”, the purpose of this paper is to provide a documentation of evaluating the quasi-global WRF-Chem simulations particularly for trans-Pacific transport, which is important and was not done in Zhao et al. (2013b) that focused on the sensitivity of modeling dust to size distributions. The text is revised as the reviewer suggested. We are following-up on using the quasi-global simulations for providing boundary conditions to higher resolution simulations to study the impacts of trans-Pacific aerosols on clouds and precipitation in the western U.S.

Specific comments:

- *The first paragraph of the introduction section discusses about the trans-Pacific transport of aerosols which has already been well known, and with much more details have been already discussed in previous articles. Seasonal variations in aerosol optical depth across the Pacific that have been later discussed in Section*

4.2.1 do not add any new insights into the current understanding of the subject. What is more important and should be discussed in the introduction and later on in the results section of the manuscript are different meteorological mechanisms that are responsible for both emission (particularly for natural aerosols such as dust) and transport of aerosols in different seasons. In this way, part of the strong seasonal variations in aerosol optical depth that have been presented in Figs. 4 and 5 can be explained. For example, as discussed by Alizadeh-Choobari et al. (2014) both shifting and strength of the prevailing wind over the Pacific Ocean are responsible for the transport pathway of aerosols and the extent that they can travel. In addition, as depending on the season, aerosols are transported at different elevations across the Pacific Ocean, the meteorological conditions behind such seasonal variations can be fully discussed. As an example, such factors for May 2007 are discussed by Uno et al. (2009).

As we stated in the manuscript, “We focus on the simulation over the trans-Pacific transport region as a first step to evaluate the simulation for providing consistent lateral chemical boundaries for nested regional simulations used to investigate the impact of transported aerosols on regional air quality and climate.” Hence the purpose of the seasonal analysis is to evaluate the quasi-global simulations of aerosols using WRF-Chem that can further provide chemical boundary conditions for nested regional simulations rather than focusing on investigating the mechanisms driving the seasonal variations of aerosols, which has been well examined in previous studies as the reviewer also mentioned. We highlighted the potential aerosol impact on the air quality and climate over the western U.S. in the introduction because the next step of research is to use this quasi-global WRF-Chem simulation to drive the nested simulations over the western U.S. for understanding the trans-Pacific aerosols’ impact there. Now we add more discussion of the previous studies about the trans-Pacific aerosols in the introduction, “Previous studies have shown that aerosols outflowed from the Asian continent can be transported by the mid-latitude prevailing westerlies across the Pacific Ocean and ultimately reach the west coast of North America and beyond, and its efficiency is the largest in spring (e.g., Takemura et al., 2002; Chin et al., 2007; Yu et al., 2008; Uno et al., 2009, 2011; Alizadeh-Choobari et al., 2014). Takemura et al. (2002) found that the contribution of anthropogenic aerosols to the total aerosol optical thickness is comparable to that of dust during the transport over the North Pacific in spring. Chin et al. (2007) found that the long-range transported dust brought 3 to 4 times more fine particles than anthropogenic pollutants to the total surface fine particles over the U.S. on annual average with a maximum influence in spring and over the northwestern U.S. Yu et al. (2008) estimated that about 25% of the Asian outflow reaches the west coast of North America, which is about 15% of the total North American emissions, and the transport fluxes are largest in spring and smallest in summer. Uno et al. (2011) also revealed that the dust trans-Pacific path sometimes could be split into two branches: a southern path to

the central U.S. and a northern path that is trapped and stagnant for a longer time and finally subsides over the northwestern U.S.”

We did briefly explain the underlying mechanisms of seasonal variation in the manuscript “Previous studies found that trans-Pacific transport of air pollutants is most efficient in MAM due to active cyclonic activity and that pollutants are lifted to the free troposphere where they can be rapidly transported across the Pacific by strong westerlies (e.g., Forster et al., 2004; Liang et al., 2004; Heald et al., 2006; Yu et al., 2008).” We also associated the model biases with meteorological processes, e.g., “The model generally underestimates the retrieved AOD over the North Pacific with the annual mean value of 0.13 that is lower than the retrieved values of 0.15 (MODIS) and 0.16 (MISR). This negative bias is mainly due to the underestimation of the oceanic AOD to the south of 20°N, which may be due to underestimation of marine emissions (Yu et al., 2003) and/or overestimation of aerosol wet removal associated with the positive bias in precipitation (Fig. 3). The discrepancy may also be due to higher uncertainty at low aerosol level (Levy et al., 2013) and cloud contamination in the retrievals that leads to an overestimation of AOD in some regions of the North Pacific (e.g., Zhang and Reid, 2006).”

- ***The averaged methods that have been used in the study caused the observed and simulated data to be missing over large areas in summer. This has led to the wrong conclusion that summer is the cleanest season in Regions 2 and 3 (lines 355 and 406, and Fig. 5), while in reality this is not the case.***

First, we sampled observations and simulations at the same time and location. Although this sampling method results in some missing data due to cloud impact on satellite retrievals, it reduces the sampling discrepancy in space and time between satellites and simulations and assures a fair comparison. Second, our results about seasonal variation of aerosols over Pacific from either satellite retrievals or simulations are generally consistent with previous studies (e.g., Fig. 3 and 4 in Yu et al., 2008, and Fig. 3 in Yu et al., 2012) using satellites and models that also show the smallest aerosol amounts in summer on average. We clarify this in the manuscript now “This seasonal variation is generally consistent with previous studies (Yu et al., 2008, 2012), although our sampling method results in more missing data from satellite retrievals in JJA than other seasons.”

Yu, H. B., L. A. Remer, M. Chin, H. S. Bian, R. G. Kleidman, and T. Diehl: A satellite-based assessment of transpacific transport of pollution aerosol, *J. Geophys. Res.*, 113, D14S12, doi:10.1029/2007JD009349, 2008.

Yu, H., L. A. Remer, M. Chin, H. Bian, Q. Tan, T. Yuan, and Y. Zhang: Aerosols from Overseas Rival Domestic Emissions over North America, *Science*, 337, 566-569, 2012.

- ***As the authors mentioned, there have been some modifications to run the WRF-Chem model on the quasi-global scale. Please briefly discuss these changes in the***

model description as this is a quite new aspect and novelty of the present study.

To configure the quasi-global WRF-Chem simulation, we modified the treatment of the chemical boundary conditions to use periodic boundary conditions in the zonal direction while the boundary treatment in the meridional direction is based on prescribed conditions. We also modified other parts of the model such as the oceanic emission schemes and convective transport scheme of tracers to produce more reasonable aerosol distributions globally. We now highlight these changes in the revised manuscript “Code modifications include changes to the chemical boundary treatment using periodic boundary conditions in the zonal direction for quasi-global WRF-Chem simulation. Other changes to the model include the oceanic (sea salt and dimethyl sulfide) emission schemes and the convective transport and removal scheme of tracers that play a significant role in quasi-global WRF-Chem simulations of aerosols.”

- ***Due to many writing problems, the manuscript should undergo a language revision.***

We made corrections to English writing.

- ***Last paragraph in page 5: The work of Alizadeh-Choobari et al. (2015) can be cited and discussed here. They conducted the WRF-Chem model to study the global distribution of mineral dust and its radiative forcing on the global scale.***

We apologize for missing the discussion of Alizadeh-Choobari et al. (2015), which is very relevant to our study. We have included it in the revision. In addition, we actually tried the WRF-Chem configuration adopted by Dr. Alizadeh-Choobari for global simulations, but we found the specific configuration only runs stably in global simulations with simple chemistry as used in Alizadeh-Choobari et al. (2015). This may be due to convergence issue of solving chemical reactions near the relatively pristine polar regions. However, more sophisticated chemistry is needed for the purpose of our studies on not only dust but also other anthropogenic aerosols. Therefore, a more stable quasi-global WRF-Chem configuration is used in our studies. We added more discussion about this in the revised manuscript “Alizadeh-Choobari et al. (2015) conducted a global WRF-Chem simulation of dust and its radiative forcing, which was configured with dust aerosol only without other aerosols and chemistry. However, WRF-Chem global simulation with sophisticated chemistry including anthropogenic and natural aerosols could not run stably due potentially to convergence issue of solving chemical reactions near the relatively pristine polar regions. Given the need of sophisticated chemistry to simulate not only dust but also other anthropogenic aerosols, a more stable near global coverage WRF-Chem configuration is used in this study to circumvent this technical difficulty to characterize the trans-Pacific transport of aerosols.”

Technical corrections:

- **Line 23: Write the WRF-Chem in full as it appears the first time in abstract.**

Done.

- **Line 25: Add “the” before “first time”**

Done.

- **Line 57 and in other parts of the manuscript: Add “the” before “Pacific Ocean”**

Done.

- **It is better to remove lines 127 to 131. The version of WRF-Chem can be mentioned in Section 2.1.**

Revised.

- **Line 147: You have mentioned that “cloud-ice-borne aerosols are not explicitly treated in the model”. Is it parameterized? Please specify that.**

Now we clarify it in the text “Cloud-ice-borne aerosols through ice nucleation of aerosols are not considered in the model, but the removal of aerosols by the droplet freezing process is considered.”

- **Line 325: Remove “of” before “2010-2014” here and throughout the manuscript.**

Done.

- **Line 338: Discuss possible explanation for the overestimation of model simulation in the specified regions.**

The overestimation of precipitation over the ITCZ may be due to biases from the convective parameterizations in producing tropical precipitation in WRF. More discussion about this is now added into the manuscript “The excessive precipitation over the tropical Pacific may be due to biases from the convective parameterizations in producing tropical precipitation in WRF, such as overestimation of precipitation efficiency from simple treatment of cloud microphysical processes in convective clouds, and biases in the prescribed temperature and humidity reference profiles (e.g., Fonseca et al., 2015; Hagos et al., 2016). Short sensitivity experiments we performed show that the WRF simulated tropical precipitation is sensitive to the choice of convective parameterizations (not shown).”

- **Line 548: You mean “the total aerosol amount”?**

We mean the total dust amount consisting of transpacific dust and local dust.

- **Line 593: remove “for first time”**

This is to emphasize that the evaluation is done for the first time, as we also state in the

abstract. Now we follow your suggestion as in abstract to change it as “for the first time”.

Anonymous Referee #2

General comments:

- *This paper presents a nice evaluation of a quasi-global WRF-Chem model in simulating trans-pacific transport of aerosol. Multiple satellites and in-situ observations are used to evaluate spatial, temporal and vertical distribution of aerosol simulations. Aerosol species over the West US and their contributions from local emissions are also discussed. This paper is well written and very useful for other researchers. I only have a few minor comments that should be addressed prior to publications.*

We thank the reviewer for a detailed review. Both text and figures are revised as the reviewer suggested.

Minor comments:

- ***L170: Could you specify what kind of technical difficulties in running global WRF-Chem?***

Before configuring the quasi-global WRF-Chem simulations, we experimented with WRF-Chem configurations for global simulations, and found that only global simulations without sophisticated chemistry as adopted by Alizadeh-Choobari et al. (2015) can run stably. The reason may be due to convergence issue of solving chemical reactions near the relatively pristine polar regions. However, sophisticated chemistry is needed to simulate not only dust but also other anthropogenic aerosols. Therefore, a more stable quasi-global WRF-Chem configuration is used in our studies. We now add more discussion about this in the revised manuscript “Alizadeh-Choobari et al. (2015) conducted a global WRF-Chem simulation of dust and its radiative forcing, which was configured with dust aerosol only without other aerosols and chemistry. However, WRF-Chem global simulation with sophisticated chemistry including anthropogenic and natural aerosols could not run stably due potentially to convergence issue of solving chemical reactions near the relatively pristine polar regions. Given the need of sophisticated chemistry to simulate not only dust but also other anthropogenic aerosols, a more stable near global coverage WRF-Chem configuration is used in this study to circumvent this technical difficulty to characterize the trans-Pacific transport of aerosols.”

- ***L283-286: Is CALIOP data collocated with model simulations? Please clarify.***

Yes. Now we clarify it in the manuscript “The model results are sampled for averaging at the locations and times where and when retrievals are available.”

- ***L387-389: What’s the mean values for model simulations?***

The mean values from the model simulation are the same as those from the AERONET data. Now we clarify it as “The model reproduces exactly these values at the three sites

with correlation coefficients of 0.45, 0.65, and 0.64, respectively.”

- ***L441: “followed by DJF”?***

Revised.

- ***Figure 15: This figure needs some improvement. The dots are not easy to see.***

Now we revised the figure responding to the comments from other reviewers. Figure 15 now shows the monthly comparison between observation and simulation. The quality is improved.

Anonymous Referee #3

General comments:

- *This manuscript presents the model evaluation of 5-year quasi-global WRF-Chem simulations using various surface and satellite observational and reanalysis datasets. Despite the lack of direct aerosol measurement data especially over East Asia and Pacific Ocean, authors are able to use available satellite or ground-based retrieved aerosol optical properties such as AOD, AAOD, and EAE to compare with simulations and draw the conclusion that WRF-Chem model can well simulate the spatial and seasonal variability of aerosol properties and transport and evolution of aerosols over the trans-Pacific domain during the 5-year time period. This manuscript is generally well written with many interesting analyses. It's definitely of scientific interest to the research community and I would recommend it to be accepted after a minor revision.*

We thank the reviewer for a detailed review. Both text and figures are revised as the reviewer suggested.

Specific comments:

- *Lines 110-111: Using in-situ observational aerosol (including dust) data to evaluate the simulations especially over pollutant source regions such as East Asia for the trans-Pacific domain is critical to demonstrate the model's capability in accurately simulating aerosol transport/evolution. There are actually a few regular networks from China, Japan, and Korea that provide long-term observational or observational-derived data for PM_{2.5} or PM₁₀ to the public. However it takes some efforts in order to collect those data and might be out of the scope of work to the authors. At least I would like to see this lack of evaluation using in-situ data to be acknowledged as a limitation in the summary section.*

We used mostly space and ground based remote sensing data to evaluate the simulations because we cannot obtain the public-released observations of PM over East Asia for the simulation period. Although we are aware of the data from the Acid Deposition Monitoring Network in East Asia (EANET), they provide measurements only up to 2008, which may not be suitable for evaluating our simulations for 2010-2014. To the best of our knowledge, there are no publicly available in-situ observations that can be used to evaluate our simulated aerosols over East Asia. We thank the reviewer for pointing out possible sources of data that may be obtained from researchers in East Asia through collaboration, which would be included in our future studies. We have acknowledged this in the introduction “For lack of in-situ observations over East Asia and the Pacific Ocean for our simulation period, evaluation is performed mainly using reanalysis and satellite retrieval (e.g., CALISPO, MODIS, and MISR) datasets, along with available ground-based observations from AERONET and IMPROVE in the region.” and in the summary, we add “Evaluation of model results with in-situ observations would be informative. In-

situ data even for specific events are valuable especially over Asia and the Pacific where public data are currently sparse or inaccessible, although some observations may be obtained through collaborations.”

- ***Lines 338-339: What may cause this large overprediction for precipitation (authors didn't show any statistics, however from the plots alone it seems that the overprediction is more than 50% for some seasons)?***

The overestimation of precipitation over the ITCZ may be due to biases in the convective parameterizations that produce a large fraction of tropical precipitation in WRF. More discussion about this is now added into the manuscript “The simulation reasonably reproduces the spatial and seasonal variations of precipitation, with spatial correlation coefficients of 0.89, 0.81, 0.81, and 0.84 for DJF, MAM, JJA, and SON, respectively. The simulation overestimates annual mean precipitation averaged over the North Pacific (3.1 mm day⁻¹ from GPCP versus 4.2 mm day⁻¹ from WRF-Chem). The overestimation (more than 50%) is particularly large over the Inter-Tropical Convergence Zone (ITCZ) and the western tropical Pacific located south of 20°N and the major pathway of trans-Pacific transport. The excessive precipitation over the tropical Pacific may be due to biases from the convective parameterizations in producing tropical precipitation in WRF, such as overestimation of precipitation efficiency from simple treatment of cloud microphysical processes in convective clouds, and biases in the prescribed temperature and humidity reference profiles (e.g., Fonseca et al., 2015; Hagos et al., 2016). Short sensitivity experiments we performed show that the WRF simulated tropical precipitation is sensitive to the choice of convective parameterizations (not shown).”

- ***Line 344: WRF-Chem provides AOD on several wavelengths, none of which are exactly 550 (for AERONET and MODIS) or 500 nm (for OMI). I am curious if any interpolation has been done to match with satellite or ground-based retrievals?***

Yes, interpolations were performed across several wavelengths. Now we clarify it in the manuscript “The WRF-Chem simulated AOD at 600 nm and 400 nm are used to derive the AOD at 550 nm (using the Angström exponent).” and “The model simulated AAOD at 600 nm and 400 nm are used to derive the AAOD at 500 nm (using the Angström exponent).”

- ***Lines 366-367: There are large discrepancies between MODIS and MISR AOD over western U.S. What are the exact causes? Could authors provide the retrieval uncertainties between two retrievals? Also it seems that high AOD values over western U.S. is collocated with some dust source regions. I would like to see some linkage between the dust performance of the model and AOD here over western U.S.***

Although the MODIS retrieval of high AOD over the western U.S. is sometimes

collocated with the dust source regions, the magnitude of AOD is significantly overestimated because of large uncertainties in the assumed surface reflectance in semi-arid regions (Remer et al., 2005; Levy et al., 2013). In comparison, the MISR observations in the western U.S. show better quality presumably because of the MISR multi-angle capability, allowing for a better characterization of surface reflectance. Now this is clarified in the manuscript “**The MODIS retrieval shows higher AOD over the semi-arid regions (e.g., Northwest China and the southwestern U.S.) than the MISR retrieval; however the MODIS retrieved AOD magnitude over these regions is significantly overestimated because of its large uncertainties in the assumed surface reflectance in semi-arid regions (Remer et al., 2005; Levy et al., 2013). In comparison, the MISR observations in the western U.S. show better quality presumably because of the multi-angle capability that allows for a better characterization of surface reflectance.**”

- ***Figure 8: It should be filled dots instead of black dots. Are there any meaning of the positioning of triangles and circles in addition to representing MODIS and WRF-Chem, since the positioning looks to me quite random?***

Corrected. The triangles and circles represent the mean values, which is clarified now.

- ***Lines 428-429: The larger EAE over West Pacific reflects smaller aerosol sizes and should be due to that large particles have been deposited (through either dry or wet deposition) during the long-range transport.***

It is true that EAE could become larger because larger particles are removed preferentially during the transport from Asia continent to West Pacific. However, in our study, EAE is reduced after long-range transport from the West Pacific to the East Pacific. The latter region should have larger particles deposited than the former one. This decrease of EAE is likely from the larger fractional contribution of sea salt particles in the latter region.

- ***Line 438: Again any interpolation here?***

Yes, we did. Now we clarify it. See the response to your previous comment.

- ***Lines 556-557: Uncertainties in the model treatment of aerosol thermodynamics/dynamics (e.g., condensation) may also significantly contribute to the nitrate biases.***

Now we revise it as “**The simulation has relatively larger positive biases (a factor of 2) in months (February, March, October, and November) between the cold and warm seasons, which may reflect model deficiency in aerosol thermodynamics (i.e., the partitioning of nitrate aerosol to the gas phase in these months is too slow in the model).**”

- ***Line 583: Are biogenic emissions from MEGAN? This information should be***

added in the model description.

Now we add “Biogenic emissions are calculated following Guenther et al. (1994).”

Technical notes:

- *Figure 5: The variation bar is out of bound in the figure for MODIS in some seasons. This should be fixed. Similar issues also occur in Figures 8 and 9.*

Corrected.

- *Line 409: higher AOD than MISR.*

Corrected.

- *Line 544: sulfate.*

Corrected.

Anonymous Referee #4

General comments:

- *This study evaluated a fully coupled meteorology-chemistry model (WRF-Chem) configured to conduct quasi-global simulation for years of 2010-2014 using multiple observation datasets. The evaluation has been focused on the simulation over the trans-Pacific transport region.*

After going through the manuscript, though a lot of analysis and comparison have been done between model and observation, I still have some concerns regarding the simulation and results, especially the relative old anthropogenic emission inventory and the way to including biomass burning emissions. Also, as an evaluation paper, I did not see to much quantitative analysis and conclusions when compare the differences between model and observation. In some places, the scientific points are not well presented, and some presentations and conclusions are more or less like an assumption description or arbitrary statement, which need to provide enough evidences or references to convince the reader.

I recommend this paper for publication in GMD after major revision if the authors satisfactorily address all the comments and questions.

We thank the reviewer for a detailed review. More clarification about the experiment set-up and quantitative analysis are added. Both text and figures are revised as the reviewer suggested. More detailed responses are provided in the following.

Specific comments:

- *Section 2.3, the emissions data is a very important input data for the part of chemical transport model. I am surprised that that the author did not use the recently updated anthropogenic emission inventory, e.g. the HTAP v2.1, which has been widely used from last year in a lot of model, including WRF-Chem. Even for the emissions over Asia and US, the MEIC (<http://www.meicmodel.org/dataset-mix.html>) and NEI 2010 are also the updated version compare to the emission inventory used in the manuscript. When the evaluated results show big differences between model and observations, how do the authors quantify how much is due to emission uncertainty and how much is due to model performance in simulating the long-range transport? So I strongly recommend the authors to use the recent anthropogenic emissions or the updated version.*

We didn't use the latest global emission dataset HTAP because the experiments were conducted before the dataset was publicly released (Janssens-Maenhout et al., 2015). However, as we described in the manuscript, the two key regions (East Asia and the U.S.) where we investigated the impact of trans-Pacific aerosol to the West U.S. are updated with recently available dataset. Now we correctly stated that the anthropogenic emissions over the U.S. are from NEI 2011. In the manuscript, we also clarified that **“Over East Asia, the Asian emission inventory described by Zhang et al. (2009) at 0.5°x0.5°**

horizontal resolution for 2006 is used except that BC, OC, and sulfate emissions over China are from the China emission inventory for 2010 described by Lu et al. (2011) at a 0.1°x0.1° horizontal spatial resolution and a monthly temporal resolution for the simulation period.” We thank the reviewer for bringing to our attention the MEIC inventory and we notice that this is also a relatively new dataset (Li et al., 2016) in review status. Li et al. (2016) also adopted the emission inventory in Lu et al. (2011) for India, and estimated Asian emission growth rates from 2006 to 2010 as follows: -8.0% for SO₂, +19% for NO_x, +4% for CO, +15% for NMVOC, +2% for NH₃, -3% for PM₁₀, -2% for PM_{2.5}, +6 % for BC, +2 % for OC. The most significant changes are for NO_x and NMVOC that exceed 10% difference. Both HTAP and MEIC emission inventories will be used in our future studies.

We now add some discussion about uncertainties associated with anthropogenic emissions in the summary “Last but not least, the model biases against observations may be also partly contributed by uncertainties in the emissions. Some recently updated anthropogenic emissions (e.g., Janssens-Maenhout et al., 2015; Li et al., 2016) and other biomass burning emissions with higher temporal and spatial resolutions (e.g., Wiedinmyer et al., 2011) may be used in future studies to investigate the impact of emission uncertainties on trans-Pacific aerosols over the West U.S.”

- *Section 2.3: I am wondering how did the authors include the GFED3 biomass burning emission into the model? Normally, the standard WRF-Chem code uses PREP- CHEM-SOURCE to generate and include the GFED3 biomass burning emission in the forecast. However, it can only generate the emission rate of the first simulation day (month) if the authors did not cycle the chemistry in the simulation everyday (every month). Otherwise, the authors should modify the code update (read) the GFED3 biomass burning every day (every month). Actually, the GFED3 has daily data, why did the author only include the monthly data when compared with the daily observation (IMPROVE data).*

The GFEDv3 monthly biomass burning emission fluxes are read in every day with our code modification. We now clarify it in the manuscript “Biomass burning emissions are obtained from the Global Fire Emissions Database, Version 3 (GFEDv3) with monthly temporal resolution (van der Werf et al., 2010) and vertically distributed following the injection heights suggested by Dentener et al. (2006) for the Aerosol Comparison between Observations and Models (AeroCom) project. The WRF-Chem code is modified to update the biomass burning emissions every day.” We now also add the discussion about uncertainties associated with emissions in the summary (see the response to the comment above). Since we focus more on evaluating the monthly, seasonal, and annual variation of transpacific aerosols in this study, the simulation results are now compared with monthly IMPROVE observations in Figure 15. More quantitative discussion is also added (see the response to other comments).

- ***Section 4, Figure 2, 3, 4: Please show the difference between model and observation, especially with quantitative presentation (e.g. percentage differences).***

We keep Figure 2 as is because the simulation captures the wind circulation quite well as we clarify now with more quantitative discussion “Strong westerly winds occur over the North Pacific throughout the seasons with a peak (up to 12 m/s; 5.48 m/s on spatial average) in boreal winter (DJF) followed by boreal spring (MAM) (4.46 m/s on spatial average). The winds are weakest in boreal fall (SON) (4.1 m/s on spatial average). In general, the model can well reproduce the spatial pattern of winds across the Pacific with wind speeds of 4.1-5.41 m/s averaged spatially for the four seasons, with a spatial correlation coefficient of 0.98 throughout the seasons.”

Figure 3 is now revised to show the difference between the simulations and observations. More quantitative discussion is also added in the manuscript “The simulation reasonably reproduces the spatial and seasonal variations of precipitation with spatial correlation coefficients of 0.89, 0.81, 0.81, and 0.84 for DJF, MAM, JJA, and SON, respectively. The simulation overestimates annual mean precipitation averaged over the North Pacific (3.1 mm day⁻¹ and 4.2 mm day⁻¹, respectively, from GPCP and WRF-Chem). The overestimation (more than 50%) is particularly over the Inter-Tropical Convergence Zone (ITCZ) and the western tropical Pacific that are south to the 20°N and the major pathway of trans-Pacific transport. The excessive precipitation over the tropical Pacific may be due to biases from the convective parameterizations in producing tropical precipitation in WRF, such as overestimation of precipitation efficiency from simple treatment of cloud microphysical processes in convective clouds, and biases in the prescribed temperature and humidity reference profiles (e.g., Fonseca et al., 2015; Hagos et al., 2016). Short sensitivity experiments we performed show that the WRF simulated tropical precipitation is sensitive to the choice of convective parameterizations (not shown)”

The quantitative comparison of Figure 4 is partly shown in Figure 5. Now we add more quantitative discussion about Figure 4 as “The WRF-Chem simulation generally well captures the observed spatial and seasonal variability of AOD across the Pacific with the spatial correlation coefficients of 0.63-0.76 for the four seasons against the MISR retrievals. The model generally underestimates the retrieved AOD over the North Pacific (0°-60°N, 120°E-120°W) with an annual mean value of 0.11, which is lower than the retrieved values of 0.14 (MODIS) and 0.15 (MISR). Over the region north of 20°N (20°N-60°N, 120°E-120°W), the simulation produces an average AOD of 0.14 that is more consistent with the retrieved values of 0.15 (MODIS) and 0.16 (MISR). This negative bias of the oceanic AOD south of 20°N may be due to underestimation of marine emissions (Yu et al., 2003) and/or overestimation of aerosol wet removal associated with the positive bias in precipitation (Fig. 3).” For the discussion of Figure 5, text has been revised as “The retrievals show clearly that AOD peaks in MAM followed by DJF in all the regions across the Pacific. The simulated annual mean AOD of 0.21,

0.16, and 0.09 over the West, Central, and East Pacific, respectively, successfully reproduce the observed values of 0.22, 0.16, and 0.10 from MODIS and 0.21, 0.16, and 0.10 from MISR. The simulation also captures the seasonal variability with the maximum AOD in MAM followed by DJF. In general, the MODIS and MISR retrievals and simulation consistently show that AOD is reduced from the West Pacific to the East Pacific.”

- ***P18, L398: The MODIS overestimation is compared to AERONET? Please quantify their differences.***

Yes, the MODIS retrievals overestimate AOD against the AERONET retrievals. More quantitative discussion is now added in the manuscript “Over East Asia, the MISR and AERONET retrievals agree well with the annual mean of 0.37 and 0.33, respectively. Their monthly variation correlates with a coefficient of 0.8. The MODIS retrievals with the annual mean of 0.48 generally overestimate AOD against the AERONET retrievals and correlate with the AERONET retrieved monthly AOD with a coefficient of 0.67. The simulation reproduces the AERONET observed AOD variability with an annual mean of 0.38 and a monthly correlation coefficient of 0.74.”

- ***P19, L403: Are there any references about the domination of sea-salt aerosol?***

Yes, Smirnov et al. (2003) also concluded that sea-salt aerosol dominates AOD over this Pacific island in the cold season. The manuscript is revised with more clarification and quantitative discussion as “Over the island of Pacific (the Midway_Island site), retrievals from AERONET, MODIS, and MISR are generally consistent with each other on annual mean with values of 0.14, 0.13, and 0.14, respectively. The MISR retrievals correlate well with the AERONET retrievals in monthly variation with a coefficient of 0.70, which is 0.42 for MODIS, showing a minimum in summer months. The simulated annual mean AOD of 0.14 well reproduces the AERONET retrieval. The model also captures the AERONET retrieved monthly variation of AOD with a correlation coefficient of 0.64. The simulation shows that this monthly variation is largely determined by the variation of sea-salt aerosol (e.g., Smirnov et al., 2003) and Asian pollutant outflow.”

- ***P19, Figure 7: Please explain the underestimate in in July and August over the West US.***

Now we add the discussion about this in Section 4.2.1 “Over the western U.S., the MISR and MODIS retrievals well capture the monthly variation of AERONET retrievals with correlation coefficients of ~0.9, but MISR and MODIS retrieve an annual mean AOD of 0.12 and 0.20, respectively, which are higher than the AERONET retrieval of 0.07, particularly in March-October. The simulated annual mean AOD of 0.07 reproduces the AERONET retrieval. The simulation also correlates well with the AERONET retrievals with a coefficient of 0.76 in monthly variation. Both the AERONET retrieval and

simulation show that the largest AOD occurs in the spring months, which has significant contribution from the dust aerosol transported across the Pacific (to be discussed in Section 4.5). The simulation compares more consistently with the AERONET retrieval than with the MISR and MODIS retrievals in terms of magnitude, which suggests that the difference between the MODIS and MISR retrievals and the simulation over the western U.S. shown in Figure 4, is largely due to uncertainties associated with the satellite retrievals. The simulation underestimates the AERONET retrieved AOD in July-September. This underestimation may come from the model significant negative biases in carbonaceous aerosols in the warm season (to be discussed in Section 4.5).” and in Section 4.5 “The observed OC still shows the peak concentration of $1.27 \mu\text{g m}^{-3}$ in JJA, and the model significantly underestimates the peak OC concentration with a value of $0.20 \mu\text{g m}^{-3}$. The negative bias of OC over the Southwest seems not to be related to the underestimation of biomass burning because BC is reasonably simulated. This seasonal variability may be determined by the secondary production of OC, which peaks in JJA because photochemistry is more active and emissions of biogenic VOCs are higher in the warm season. The underestimation of secondary organic aerosol (SOA) may be due to uncertainty of biogenic emissions (Zhao et al., 2016) and the outdated SOA mechanism used in the current version of WRF-Chem (Shrivastava et al., 2011).”

- ***P20, L442-444: Any explanations about this conclusion?***

More quantitative discussion and explanation are added “The simulated seasonal mean AAOD of 0.015 over the West Pacific agrees reasonably well with the OMI retrieval of 0.014 in DJF but is higher in the other three seasons, with the largest difference in JJA. The significantly lower AAOD in seasons other than DJF from the OMI retrieval is also shown in the comparison with the AERONET retrieval (to be discussed with Fig. 10). Over the Central Pacific, the simulated seasonal mean AAOD of 0.014 and 0.006 in MAM and SON, respectively, generally reproduces the retrieved AAOD of 0.017 and 0.005, but the model overestimates (underestimates) the retrieved values in JJA (DJF) with 0.008 (0.005) from the simulation and 0.004 (0.009) from the retrieval. This difference may reflect the model deficiency in simulating Asian BC outflow over the Pacific in JJA and DJF, but may also result from retrieval uncertainties. The OMI retrievals may have difficulty in distinguishing the ocean color effects from those of low aerosol concentrations in the UV spectral range and ignoring the less-sufficient amounts of absorbing aerosols (Veihelmann et al., 2007; Torres et al., 2013). Jethva et al. (2014) found that the most important source of uncertainty in OMI AAOD is the effect of sub-pixel cloud contamination related to the sensor’s coarse spatial resolution, which causes AAOD underestimations for cases of low aerosol load. Over the East Pacific, the simulated seasonal mean AAOD of 0.0035, 0.0091, 0.0048, and 0.0042 for DJF, MAM, JJA, and SON, respectively, are generally consistent with the retrieved values of 0.005, 0.007, 0.0012, and 0.003, which shows the maximum value in MAM. The most

significant difference occurs in JJA. Similar as over the Central Pacific, the underestimation of retrieved AAOD over the clean region may contribute to the difference. The retrievals and simulation show large variability of AAOD, and they generally agree within the 10th and 90th percentiles of each other. AAOD is larger over the West Pacific than the Central and East Pacific, which is consistent with the AOD pattern. The simulation shows that AAOD peaks in MAM followed by JJA over the three sub-regions, which may be due to the stronger outflow of dust and anthropogenic pollutants in the two seasons.”

- ***L444-447: I don't think so. I did not see that the model is able to reproduce the seasonal variation well. Again, please provide quantitative value using statistic method to convince the readers.***

The manuscript is revised with more quantitative discussion and explanation. See the response to the comment above.

- ***L468-L470, the seasonal variation of anthropogenic BC emission overs Asian is not such significant to make this big differences, which can be found in either INTEX-B or MEIC emission inventory.***

The manuscript is revised as “The simulation generally captures the observed monthly variability with the minimum AAOD of 0.035 and 0.032 in July from the simulation and the AERONET retrieval, respectively, and the maximum of 0.055 and 0.054 in October, respectively. The model overestimates AAOD in the warm months (May-September) with the mean values of 0.046 and 0.036 from the simulation and retrieval, respectively, and underestimates AAOD in December and January with the mean values of 0.037 and 0.043, respectively. The model positive (negative) biases in AAOD in the warm (cold) months may be partly related to the constant anthropogenic BC emissions applied throughout the seasons, but previous studies have shown that anthropogenic BC emissions over China may have seasonal variation, with roughly 6% versus 13% of the annual total BC emission in summer and winter, respectively, estimated in Lu et al. (2011).”

- ***L471-474: I don't understand the point.***

It is revised as “The lower OMI AAOD over East Asia may also indicate its negative biases over the West Pacific (Fig. 9) where the air is significantly affected by the East Asian outflow. The biases in the OMI algorithm of retrieving SSA over East Asia may be also applied over the West Pacific.”

- ***Figure 11: there are big differences if the authors quantify them, especially under 1km. It is not such subjective to get this conclusions.***

We revised the manuscript with more quantitative discussion about the comparison as

“The model generally reproduces the aerosol extinction vertical variation with correlation coefficients of 0.95-0.97. The simulated aerosol extinction coefficients are consistent with the retrievals around 0.5-1 km with difference within 15%. The difference increases in the free troposphere and below 0.5 km. The simulation is higher than the retrieval in the free troposphere (e.g., about a factor of 2 around 4 km), which may be due to the reduced sensitivity of CALIOP to tenuous aerosol layers above 4 km (Yu et al., 2010). The lower (up to 30% lower) simulated extinction coefficients below 0.5 km in all three sub-regions may indicate negative biases in estimating marine aerosol emissions and excessive wet scavenging of the model, as shown in Fig. 4. The in-situ measurements over the region are needed for further validating both remote sensing data and the simulation.”

- ***L496: Is it Figure 11?***

It should be Figure 12 that shows the simulated vertical distributions of aerosol mass and its composition fraction.

- ***Figure 12: Any references to show similar vertical distribution?***

We are not aware of any references over this investigated region.

- ***L502-507: Which is the major factor, the retrieval bias in observation data of CALIPSO or the emission uncertainty in the inventory? This conclusion looks like assumption without any strong evidence support.***

Without in-situ measurements that are generally more accurate than remote sensing data, it is difficult to further validate the simulation and the CALIOP retrieval. We now clarify this in the manuscript as “In-situ measurements over the region are needed for further validating both remote sensing data and the simulation.” In addition, we revised the manuscript with more quantitative discussion about the comparison (see the response to the comment above).

- ***Section 4.5: Please use the general model evaluation method or figure (Taylor diagram) to provide quantitatively values before getting the conclusions.***

Figure 15 is now revised to show comparison between the monthly values from the simulation results and the IMPROVE observations. Section 4.5 is now significantly revised with more quantitative discussion and explanation of the comparison.

- ***L570: It is better to compare the GFED3 with other biomass burning emission, e.g. FINN, or refer the published results to get this conclusion. Also, about the underestimate of BC and OC, please see my comment 2 about including biomass burning into the model.***

We modified the WRF-Chem code to read in the GFEDv3 biomass burning emissions

daily (see our response to your comments above). We also compare the GFED and FINN emission inventories. Now we add more discussion in the manuscript “At the Northwest sites, the observed BC and OC show significant seasonal variation with the highest surface concentration in June-September (JJAS). The sensitivity simulation shows that the peak is dominated by the North American emission that is contributed by biomass burning with a maximum in JJAS (Chin et al., 2007). The simulation captures this seasonality to some extent with monthly correlation coefficients of 0.74 and 0.69 for BC and OC, respectively. However, the simulation significantly underestimates the JJAS peak with $0.05 \mu\text{g m}^{-3}$ and $0.49 \mu\text{g m}^{-3}$ BC and $0.5 \mu\text{g m}^{-3}$ and $4.5 \mu\text{g m}^{-3}$ OC from the simulation and retrieval, respectively. This significant negative bias in the model is likely from uncertainties in the GFEDv3 biomass burning inventory for the simulation period. The monthly mean emissions at a relatively coarse horizontal resolution may not be able to capture the strong local fire events. Mao et al. (2011) pointed out that the GFED inventory may underestimate the magnitude of biomass burning emissions in the western U.S. due to the issue of detecting small fires, for example, from prescribed and agricultural burning (e.g., Randerson et al., 2012; Giglio et al., 2010). Mao et al. (2014) estimated that the biomass burning BC emissions inverted from the IMPROVE observations can be a factor of 5 higher than the GFED inventory in July-September over the Western U.S.. Another biomass burning emission inventory FINN (Fire INventory from Ncar) (Wiedinmyer et al., 2011) also shows a factor of 3 higher BC emissions than the GFED inventory over the Northwest U.S. (100°W - 125°W and 40°N - 50°N) in September 2011 (not shown).”

We also add the discussion about uncertainties associated with emissions in the Summary section “Last but not least, the model biases against observations may be also partly contributed by the uncertainties in emissions. Some recently updated anthropogenic emissions (e.g., Janssens-Maenhout et al., 2015; Li et al., 2016) and other biomass burning emissions with higher temporal and spatial resolutions (e.g., Wiedinmyer et al., 2011) may be used in future studies to investigate the impact of emission uncertainties on trans-Pacific aerosols over the West U.S.”