Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

Trans-pacific transport and evolution of aerosols: Evaluation of quasi-global WRF-Chem simulation with multiple observations

[1,2]Zhiyuan Hu, [2]Chun Zhao, [1]Jianping Huang, [2]L. Ruby Leung, [2]Yun Qian, [3,4]Hongbin Yu, [5]Lei Huang, [5]Olga V. Kalashnikova

[1]Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, Lanzhou University, Gansu, China

[2]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA

[3]Earth System Science Interdisciplinary Center, University of Maryland, MD, USA

[4]Earth Science Division, NASA Goddard Space Flight Center, MD, USA

[5]Jet Propulsion Laboratory, NASA, Pasadena, CA, USA

Manuscript for submission to *WRF-Chem special issue in Geosci. Model Dev.*

*Corresponding authors:

Chun Zhao, phone: (509) 371-6372; email: chun.zhao@pnnl.gov

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

Abstract

A fully coupled meteorology-chemistry model (WRF-Chem) has been configured to conduct quasi-global simulation for the 5 years of 2010-2014 and evaluated with multiple observation datasets for first time. The evaluation focuses on the simulation over the trans-Pacific transport region using various reanalysis and observational datasets for meteorological fields and aerosol properties. In general, precipitation and winds are well simulated by the model. The simulation captures the overall spatial and seasonal variability of satellite retrieved aerosol optical depth (AOD) and absorbing AOD (AAOD) over the Pacific that is determined by the outflows of pollutants and dust and the emissions of marine aerosols. The assessment of simulated extinction Angstrom exponent (EAE) indicates that the model generally reproduces the variability of aerosol size distributions as seen by satellites. In addition, the vertical profile of aerosol extinction and its seasonality over the Pacific that are dominated by marine aerosols near the surface and the outflow of pollutants and dust above 4 km are also well simulated. The difference between the simulation and satellite retrievals can be mainly attributed to model biases in estimating marine aerosol emissions as well as the satellite sampling and retrieval uncertainties. Compared with the surface measurements over the western U.S., the model reproduces the observed magnitude and seasonality of dust, sulfate, and nitrate surface concentrations, but significantly underestimates the peak surface concentrations of carbonaceous aerosol likely due to model biases in the spatial and temporal variability of biomass burning emissions and secondary organic aerosol (SOA) production. A sensitivity simulation shows that the trans-Pacific transported dust, sulfate, and nitrate can make significant contribution to surface concentrations over the rural areas of the

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

western U.S., while the peaks of carbonaceous aerosol surface concentrations are dominated by the North American emissions. Both the retrievals and simulation show small interannual variability of aerosol characteristics for 2010-2014 averaged over three Pacific sub-regions. The evaluation in this study demonstrates that the WRF-Chem quasi-global simulation can be used for investigating trans-Pacific transport of aerosols and providing reasonable inflow chemical boundaries for the western U.S. to further understand the impact of transported pollutants on the regional air quality and climate with high-resolution nested regional modeling.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

1 Introduction

Aerosols, including from natural and anthropogenic sources in Europe, North Africa, and East Asia, can be transported across Pacific Ocean thousands of miles downwind to North America and even beyond. Previous studies using ground-based and satellite measurements and numerical models estimated about 7-10 days of travel time for aerosols to traverse the Pacific Ocean (Eguchi et al., 2009). These transported aerosols can play an important role in atmospheric composition (Yu et al., 2008), air quality (Jaffe et al., 1999; VanCuren, 2003; Heald et al., 2006; Chin et al., 2007; Fischer et al., 2009; Yu et al., 2012; Tao et al., 2016), and regional weather and climate (Eguchi et al., 2009; Yu et al., 2012; Creamean et al., 2013; Fan et al., 2014) over the U.S. West Coast. At the surface, Heald et al. (2006) found that Asian anthropogenic aerosol plume increased aerosol concentration in elevated regions of the northwestern U.S. by 0.16 μg m^{-3} in spring 2001. Chin et al. (2007) also found that long-range transported dust increased the annual mean fine particle concentrations by 0.5-0.8 μg m^{-3} over the western U.S., with a maximum enhancement in spring. The trans-Pacific transported aerosols can also significantly absorb and scatter solar radiation (Yu et al., 2012; Fast et al., 2014; Tao et al., 2016), and serve as cloud condensation nuclei and ice nuclei that affect winter storms in the western U.S. (Sassen, 2002; Ault et al., 2011; Creamean et al., 2013; Fan et al., 2014). Deposition of the transported aerosols on snowpack in elevated regions (Hadley et al., 2010) may also accelerate snowmelt and influence the regional hydrological cycle and climate over the western U.S. (Qian et al., 2009 and 2015; Painter et al., 2010). Hence it is important to quantify the trans-Pacific transport of aerosols and how they evolve over the long distance.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

Previous studies have used global models to quantify the long-range transport of aerosols to the western U.S. (Fairlie et al., 2007; Heald et al., 2006; Chin et al., 2007; Hadley et al., 2007). However, simulations were performed at relatively coarse resolutions (typically 1-2 degrees) that cannot fully resolve the large geographical variability of aerosols over the western U.S. with complex topography (Zhao et al., 2013a). Coarse resolution simulations also lack the capability to fully resolve aerosol-cloud-precipitation interaction. Some studies have reported regional simulations at relatively high resolutions over the western U.S. (e.g., Zhao et al., 2013a; Fan et al., 2014; Fast et al., 2014). However, most of them either used sparse in-situ observations to provide lateral boundary conditions that are only suitable for idealized or short-term sensitivity studies, or used simulations from global models with inconsistent physics and chemistry schemes to provide lateral boundary conditions, which introduce biases in estimating the contribution and effect of trans-Pacific transported aerosols.

To investigate the impact of trans-Pacific transported aerosols on regional air quality and climate of the US West Coast, a multi-scale modeling framework including global simulation at coarse resolutions that captures the large-scale circulation and provide consistent chemical lateral boundaries for nested regional simulation at high resolutions is needed. WRF-Chem, the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) coupled with a chemistry component (Grell et al., 2005), is such a modeling framework. As a state-of-the-art model, WRF-Chem supports nested simulations, and includes complex aerosol processes and interactions between aerosols and radiation, clouds, and snow albedo (Zhao et al., 2014). The model has been used extensively to study aerosols and their impacts on air quality and climate at regional

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

scales (e.g., Fast et al., 2006, 2009; Gustafson et al., 2007; Qian et al., 2010; Gao et al., 2011, 2014; Shrivastava et al., 2011; Chen et al., 2013, 2014; Zhao et al., 2010, 2011, 2012, 2013a; 2014). Zhao et al. (2013b) is the first study to use WRF-Chem for quasi-global (180° W-180° E, 60° S-70° N) simulations at a resolution of 1° × 1° to examine uncertainties in simulating global dust mass balance and radiative forcing.

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. In this study, the WRF-Chem simulation for 2010-2014 is evaluated extensively using observational data. For lack of in-situ observations over East Asia and Pacific Ocean during the simulation period, evaluation is performed mainly using reanalysis and satellite retrieval (e.g., CALISPO, MODIS, and MISR) datasets, along with some available ground-based observations from AERONET and IMPROVE in the region. 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. Spatial evolution of aerosols during the trans-Pacific transport as well as their seasonal and annual variability simulated by WRF-Chem will also be characterized.

In the following sections, the detailed setup of WRF-Chem will be described in Section 2. In Section 3 ground-based measurements and satellite retrievals will be presented. In Section 4, we evaluate the WRF-Chem simulated spatial distributions and

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

seasonal and annual variability of aerosols across the Pacific with the observations. The conclusion can be found in Section 5.

2 Model description

In this study, WRF-Chem (3.5.1), updated by scientists at Pacific Northwest National Laboratory (PNNL), is used and briefly described in Section 2.1. Section 2.2 discusses the setup of model simulations for this study. In Section 2.3, the emissions used in the simulations are described, including anthropogenic and biomass burning emissions, and mineral dust and sea-salt emissions.

2.1 WRF-Chem

The MOSAIC (Model for Simulation Aerosol Interactions and Chemistry) aerosol module (Zaveri et al., 2008) coupled with the CBM-Z (carbon bond mechanism) photochemical mechanism (Zaveri and Peters, 1999) in WRF-Chem is selected in this study. MOSAIC uses a sectional approach to represent aerosol size distributions with four or eight discrete size bins in the current version of WRF-Chem (Fast et al., 2006). All major aerosol components including sulfate (SO_4^{-2}), nitrate (NO_3^-), ammonium (NH_4^+), black carbon (BC), organic matter (OM), sea-salt, and mineral dust are simulated in the model. The MOSAIC aerosol scheme includes physical and chemical processes of nucleation, condensation, coagulation, aqueous phase chemistry, and water uptake by aerosols. Dry deposition of aerosol mass and number is simulated following the approach of Binkowski and Shankar (1995), which includes both turbulent diffusion and gravitational settling. Wet removal of aerosols by grid-resolved stratiform clouds and precipitation includes in-cloud removal (rainout) and below-cloud removal (washout) by

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

impaction and interception, following Easter et al. (2004) and Chapman et al. (2009). Cloud-ice-borne aerosols are not explicitly treated in the model, but the removal of aerosols by the droplet freezing process is considered. Convective transport and wet removal of aerosols by cumulus clouds follow Zhao et al. (2013b).

Aerosol optical properties such as extinction, single scattering albedo (SSA), and asymmetry factor for scattering are computed as a function of wavelength for each model grid box. Aerosols are assumed internally mixed in each bin (i.e., a complex refractive index is calculated by volume averaging for each bin for each chemical constituent of aerosols). The Optical Properties of Aerosols and Clouds (OPAC) data set (Hess et al., 1998) is used for the shortwave (SW) and longwave (LW) refractive indices of aerosols, except that a constant value of 1.53+0.003i is used for the SW refractive index of dust following Zhao et al. (2010, 2011). A detailed description of the computation of aerosol optical properties in WRF-Chem can be found in Fast et al. (2006) and Barnard et al. (2010). Aerosol radiative feedback is coupled with the Rapid Radiative Transfer Model (RRTMG) (Mlawer et al., 1997; Iacono et al., 2000) for both SW and LW radiation as implemented by Zhao et al. (2011). The optical properties and direct radiative forcing of individual aerosol species in the atmosphere are diagnosed following the methodology described in Zhao et al. (2013a). Aerosol-cloud interactions were included in the model by Gustafson et al. (2007) for calculating the activation and resuspension between dry aerosols and cloud droplets.

2.2 Numerical experiments

Following Zhao et al. (2013b), we use a quasi-global channel configuration with periodic boundary conditions in the zonal direction and 360 × 145 grid cells (180° W-180°

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

E, 67.5° S-77.5° N) to perform simulation at 1° horizontal resolution over the period of 2010-2014. This circumvents some technical difficulties in running global WRF-Chem in v3.5.1 for a near global coverage to characterize the trans-Pacific transport of aerosols. The simulation is configured with 35 vertical layers up to 50 hPa. The meteorological initial and lateral meridional boundary conditions are derived from the National Center for Environmental Prediction final analysis (NCEP/FNL) data at 1° horizontal resolution and 6 h temporal intervals. The modeled wind components u and v and atmospheric temperature are nudged towards the NCEP/FNL reanalysis data throughout the domain with a nudging timescale of 6 h in all cases (Stauffer and Seaman, 1990). This provides a more realistic simulation of large-scale circulation, which is important for modeling long-range transport. The chemical initial and meridional boundary conditions are taken from the default profiles in WRF-Chem, which are the same as those used by McKeen et al. (2002) and are based on averages of mid-latitude aircraft profiles from several field studies over the eastern Pacific Ocean. The impact of chemical boundary conditions on the simulated results is negligible (Zhao et al. 2013b). This study uses a set of selected schemes for model physics, including the MYJ (Mellor–Yamada–Janjic) planetary boundary layer scheme, Noah land surface scheme, Morrison 2-moment microphysics scheme, Kain-Fritsch cumulus scheme, and RRTMG longwave and shortwave radiation schemes.

2.3 Emissions

Anthropogenic emissions are obtained from the REanalysis of the TROpospheric (RETRO) chemical composition inventories (http://retro.enes.org/index.shtml) except over East Asia and the United States. Over the U.S., the National Emission Inventory

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

(NEI) 2005 (WRF-Chem user guide from http://ruc.noaa.gov/wrf/WG11/Users_guide.pdf) is used. 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. 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. Sea-salt emission follows Zhao et al. (2013a), which is based on Gong (2003) to include correction of particles with radius less than 0.2 μm and Jaegle et al. (2011) to include the sea-salt emission dependence on sea surface temperature. Vertical dust emission fluxes are calculated with the Goddard Chemical Aerosol Radiation Transport (GOCART) dust emission scheme (Ginoux et al., 2001), and the emitted dust particles are distributed into the MOSAIC aerosol size bins following a theoretical expression based on the physics of scale-invariant fragmentation of brittle materials derived by Kok (2011). For MOSAIC 8-bin, dust particles are emitted into eight size bins with mass fractions of 10^{-6}%, 10^{-4}%, 0.02%, 0.2%, 1.5%, 6%, 26%, and 45%, respectively. Although the main purpose of this study is to evaluate the WRF-Chem simulations, a sensitivity simulation, in which dust, fire, and anthropogenic emissions over North America (10°N-70°N and 170°W-60°W) are removed, is also conducted to understand the contribution of trans-Pacific transported aerosols to the surface aerosol concentrations over the western U.S.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

3 Aerosol Observations

3.1 Satellite Retrievals

3.1.1 MODIS

The Moderate Resolution Imaging Spectroradiometer (MODIS) instrument onboard the NASA EOS Terra satellite observes Earth in 36 spectral bands from 0.4 to 14.4 μm, and provides nearly daily global coverage with local equatorial overpass time of about 10:30 am since 2000 (King et al., 1999). The "dark target" algorithm has been developed to retrieve AOD and size parameters (Angstrom exponent, effective radius, and fine-mode fraction) over waters and vegetated lands (Kaufman et al., 1997; Remer et al., 2005). The "deep blue" algorithm has been implemented to retrieve AOD over bright land initially, which then has also been extended to vegetated land (Hsu et al., 2006, 2013). MODIS aerosol products have been widely used to characterize the regional, seasonal, and global distribution of aerosol and its components (Yu et al., 2003, 2009; Chin et al., 2004; Kaufman et al., 2005a), estimate aerosol radiative forcing (Yu et al., 2004; Remer and Kaufman, 2006), and study aerosol-cloud interactions (Kaufman et al., 2005b; Koren et al., 2005; Yu et al., 2007). In this study, MODIS data from the collection 5.1 are used. We use the "deep blue" AOD over land and the "dark target" AOD over ocean, both at 550 nm and at 1° x 1° horizontal resolution. Also, we use the "dark target" over-ocean extinction Angstrom exponent (EAE) over the 470-660 nm wavelength range to evaluate model simulations of particle size information (Anderson et al., 2005; Remer et al., 2005; Levy et al., 2013).

3.1.2 MISR

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

The Multi-angle Imaging SpectroRadiometer (MISR) instrument onboard the Terra spacecraft crosses the equator at ~10:30 AM local time since 1999. It observes continuously in four narrow spectral bands centered at 446, 558, 672 and 866 nm using nine separate cameras oriented along the orbital track with surface viewing zenith angles ranging from ±70.5° (Diner et al., 1998). Aerosol retrievals are performed on 16 × 16 patches of 1.1 km sub-regions, yielding an aerosol product at 17.6 ×17.6 km spatial resolution, referred to as a "Level 2" product (Martonchik et al., 2002). MISR Level 2 aerosol products have been described in Kahn et al (2009). The latest version (Version 22) of MISR aerosol product also provides the fraction of AOD due to "fine" (particle radii <0.35 μm), "medium" (particle radii between 0.35 and 0.7 μm) and "large" (particle radii >0.7 μm) particles as well as the fraction of AOD due to "spherical" and "nonspherical" particles at the four MISR spectral bands. Here, we compare the MISR AOD at 550 nm from Version 22 of Level 2 with the model results.

3.1.3 OMI

OMI onboard the NASA Aura satellite has a daily global coverage, and crosses the equator at 1:45 PM local time. The nadir horizontal resolution of OMI is 24×13 km^2. In this study the OMAERUV Level 2 Collection 003 V1.4.2 product (Jethva et al., 2014) is used as an independent data set providing SSA that is derived based on the reflectances measured by the OMI instrument at 0.39 μm. The information on aerosol absorption in OMI measurements comes, to a large extent, from the interaction with Rayleigh scattering in the UV spectral region (Torres et al., 2013). The retrieved parameters are also reported at 0.38 μm and 0.50 μm. Current OMI AOD has positive biases likely due to a combination of factors including cloud contamination, surface albedo effects,

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

radiometric calibrations, and misidentified aerosol type (Ahn et al., 2008). Therefore, in this study, OMI AAOD at 500 nm is reconstructed using the WRF-Chem simulated 500 nm AOD and OMI SSA at 500 nm with the formula of $AAOD_{OMI}=AOD_{MODEL}\times(1-SSA_{OMI})$.

3.1.4 CALIPSO

In this study, we use aerosol extinction profiles retrieved by the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite. The CALIPSO satellite was launched into a Sun-synchronous orbit on 28 April 2006. CALIOP is a dual-wavelength polarization lidar and is designed to acquire vertical profiles of attenuated backscatter from a near nadir-viewing geometry during both day and night phase (Winker et al., 2007). In this study, the aerosol extinction profiles at a nominal horizontal resolution of 5 km from the CALIPSO Level 2 profile products are used to evaluate the model. We focus on the CALIOP nighttime observations in cloud-free condition, because nighttime observations have higher accuracy than daytime observations (Winker et al., 2009).

The cloud-aerosol discrimination (CAD) score, which is an indicator that measures confidence level of the discrimination between clouds (positive value) and aerosols (negative value), is used to help screen out aerosol profiles that contain cloud signals. We include the aerosol data with CAD score between -20 and -100, the same as Yu et al., (2010, 2015). The other screening is to exclude aerosol layers where the retrieval algorithm has to adjust the initially selected lidar ratio that is based on the type and subtype of the aerosol layer to be analyzed. It should be noted that when the aerosol

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

extinction is not detected by CALIOP, we set its value and also the corresponding model result equal to zero, and then we compare CALIOP extinction profiles with the model and analyze the seasonal variation.

3.2 Ground-based observations

3.2.1 AERONET

The AErosol RObotic NETwork (AERONET) is a globally distributed remote sensing network for aerosol monitoring from ground stations (Holben et al., 1998). AERONET uses the Cimel sun/sky photometer that measures AOD in 16 spectral channels (340-1640 nm). The measurements provide products every 15 minutes during daytime. In addition, an inversion algorithm is used for the retrieval of aerosol size distribution, complex refractive index, single-scattering albedo, and phase function (Dubovik and King, 2000; Dubovik et al., 2002). The spectral AOD from AERONET has an accuracy of ±0.01 (Eck et al., 1999; Holben et al., 2001). In the analysis presented in this paper, the cloud screened and quality assured level 2.0 products are used. AERONET products do not measure at wavelength 0.55 μm, so we calculate them through using the Angstrom exponent and the values at two nearest wavelengths 0.5 μm and 0.675 μm. The AERONET sites are located along the trans-Pacific transport pathways, so the products are important for evaluating the model results. Five sites over East Asia, one island site over North Pacific, and four sites over the western U.S. are selected for comparison as shown in Figure 1.

3.2.2 IMPROVE

The Interagency Monitoring for Protected Visual Environments (IMPROVE) network was initiated in 1985 by U.S. federal agencies including EPA, National Park

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

Services, Department of Agriculture-Forest Service, and other land management agencies as a part of the EPA Regional Haze program (Malm et al., 1994). The network monitors the visibility conditions and changes in national parks and wilderness areas on a long-term basis. The detail sample collection and analytical methodology have been given by Hand et al., (2011), and the data can be downloaded from (http://views.cira.colostate.edu/fed/DataWizard/Default.aspx). There are 15 sites (Fig. 1) along the west coast selected to compare with the surface aerosols of the model. In this study, the mass concentrations of sulfate, nitrate, EC, OC, and dust in $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm) are used to evaluate the model. The fine dust was calculated following the formula (Malm et al., 1994; Zhao et al., 2013a):

$$PM_{2.5}\text{-Dust} = 2.2[Al] + 2.49[Si] + 1.63[Ca] + 2.42[Fe] + 1.94[Ti]$$

Where [Al], [Si], [Ca], [Fe], and [Ti] represent the mass concentration of aluminum, silicon, calcium, iron, and titanium, respectively.

4 Results

4.1 Wind fields and precipitation

Winds and precipitation are two crucial meteorological factors playing important roles in aerosol emission, transport, and removal. The seasonal mean wind fields at 850 hPa averaged for the period of 2010-2014 from the WRF-Chem simulation are compared with the Modern-Era Retrospective analysis for Research and Applications (MERRA) reanalysis data (Rienecker et al., 2011) (Figure 2). Strong westerly winds occur over the North Pacific throughout the seasons with a peak (up to 12 m/s) in boreal winter (DJF) followed by boreal spring (MAM). In general, the model can well reproduce the spatial

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

pattern of winds across the Pacific and its seasonality. Figure 3 shows the spatial distribution of seasonal mean precipitation from the Global Precipitation Climatology Project (GPCP) observation (Huffman et al., 2001) and WRF-Chem simulation averaged for the period of 2010-2014. Over East Asia, precipitation reaches a maximum during the boreal summer (JJA) followed by MAM. In the North Pacific basin, the largest precipitation occurs in DJF along the storm tracks with the maximum westerlies. Over the U.S. west coast, precipitation peaks during DJF and reaches a minimum in JJA. The simulation reasonably reproduces the spatial and seasonal variation of precipitation, with overestimation in some regions, particularly over the Inter-Tropical Convergence Zone (ITCZ) and western tropical Pacific that are south of the major pathway of trans-Pacific transport.

4.2 Aerosol optical depth

4.2.1 Spatial and temporal variation

Figure 4 shows the spatial distributions of seasonal mean AOD at 550 nm across the Pacific from Asia to North America averaged for 2010-2014 from the retrievals of MODIS and MISR onboard Terra and the corresponding WRF-Chem simulation. In order to reduce the sampling discrepancy between the two retrievals, the daily results from the two satellite retrievals and simulation are sampled and averaged at the same time and location. This way of averaging leads to the blank areas of missing values, which are relatively large in JJA. Satellite retrievals show that AOD is high over the Asian continent and gradually decreases across the Pacific. High AOD coincides with the sub-tropical jet (30°N-50°N, Fig. 2) over the Pacific and results from wind-induced increase

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

in sea-salt loading and the Asian pollutant outflow. Seasonal variation of aerosols across the Pacific is evident, with peak AOD over the western Pacific in MAM and minimum AOD in JJA and SON. 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 it 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). The WRF-Chem simulation generally well captures the observed spatial and seasonal variability of AOD across the Pacific. The model underestimates the oceanic AOD to the south of 20°N, which may be due to underestimation of marine emissions and/or overestimation of aerosol wet removal associated with the positive bias in precipitation. The discrepancy may also be due to cloud contamination in the retrievals that leads to an overestimation of AOD in some regions of North Pacific (e.g., Zhang and Reid, 2006). The model also simulates lower AOD over the continent of North America compared with satellite retrievals. The difference between the simulation and retrievals may be due to the uncertainty in satellite retrievals over the continents (e.g., Liu et al., 2004; Levy et al., 2010).

Since this study focuses on the trans-Pacific transport and evolution of aerosols, the Pacific is further divided into three sub-regions (Region 1: 20°N-50°N and 120°E-140°E; Region 2: 20°N-50°N and 140°E-140°W; Region 3: 20°N-50°N and 140°W-120°W) representing the West Pacific, the Central Pacific, and the East Pacific shown as the black boxes in Figure 4 for analysis. Figure 5 shows the seasonal mean 550 nm AOD over the three sub-regions from the MISR and MODIS retrievals and the corresponding WRF-Chem simulation at the pass time of MISR and MODIS, respectively, averaged for

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

2010-2014. It shows clearly that AOD peaks in MAM in all the regions across the Pacific. The simulation successfully reproduces the observed seasonal variability. In general, the MODIS and MISR retrievals are consistent and show that AOD reduces from West Pacific to East Pacific, which is also indicated by the simulation. The interannual variability of AOD over the three sub-regions is small for 2010-2014 indicated by the retrievals and simulation (not shown).

Available observations from several AERONET sites (Fig. 1) over East Asia, the Pacific, and western US are also compared with the model simulation. Figure 6 shows the comparison of observed and simulated AOD at three representative AERONET sites for 2010-2014 over East Asia, an island of Pacific, and the western US coast. The observations and simulation agree well at all three sites, and both reflect the AOD gradient from East Asia to the western U.S. as shown in Figure 4. Observed AOD is the highest with a mean value of 0.31 at the SACOL site over East Asia and reduces to 0.075 at the Midway_Island site, and 0.045 at the Frenchman_Flat site. The model reproduces these values at the three sites with correlation coefficients of 0.45, 0.75, and 0.63, respectively. About 90% of simulated AOD is within a factor of 2 of the AERONET measurements.

Figure 7 further shows the monthly variation of AOD averaged at the AERONET sites over East Asia, the Pacific island, and the West US (as shown in Fig. 1) from the AERONET observations, MODIS and MISR retrievals, and WRF-Chem simulation. For the simulated AOD, contributions by dust, BC, OC, sulfate, and other aerosols are also shown. Over East Asia, the MISR and AERONET retrievals agree well in monthly variation. The MODIS retrievals generally overestimate AOD. The simulation reproduces

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

the AERONET observed AOD variability. Model results show that anthropogenic aerosols dominate the AOD from summer to winter while dust can significantly contribute to the AOD in spring. Over the island of Pacific (the Midway_Island site), retrievals from AERONET, MODIS, and MISR are consistent with each other and with the simulation on monthly variation of AOD, primarily determined by the sea-salt aerosol, which shows a minimum in summer months. The trans-Pacific transported aerosols (other than sea-salt) also show strong monthly variation with a maximum in April and a minimum in July. Over the western U.S., both retrievals and simulation show 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). MODIS retrieved lower AOD than MISR, and both retrievals are significantly higher than that from the AERONET retrieval in March-October, while the simulation is more consistent with the AERONET retrievals, which suggests that the difference between the MODIS and MISR retrievals and the simulation over the western US shown in Fig. 4 is due to uncertainty in satellite retrievals.

4.2.2 Wavelength dependence

The wavelength dependence of AOD that can be represented by the extinction Angstrom exponent (EAE) is an indicator of aerosol particle size (Angstrom, 1929; Schuster et al., 2006). In general, relatively small values of EAE indicate that aerosol size distributions are dominated by coarse aerosols typically associated with dust and sea-salt, while relatively large values of EAE indicate fine aerosols usually contributed by anthropogenic pollution and biomass burning. Figure 8 shows the seasonal mean EAE

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

averaged for 2010-2014 from the MODIS retrievals and the WRF-Chem simulation over the three sub-regions. The retrievals show clearly that the EAE values peak in JJA and reach a minimum in DJF in all three sub-regions. This seasonality reflects the fact that photochemistry is most active in JJA to produce fine aerosol particles such as sulfate. In general, the simulation successfully reproduces the observed EAE seasonality. The retrievals and simulation also show that the values of EAE are greater in the West Pacific than in the Central and East Pacific. This pattern may reflect the dominance of the Asian pollutant outflow on the aerosol size distributions over the West Pacific, while the relatively large-size particles of sea-salt dominates in the other two regions. Again, the annual variability of EAE over the three sub-regions is small (not shown).

4.3 Aerosol absorption optical depth

Light absorbing aerosols such as BC and dust play an important role in the atmosphere to absorb radiation and change the heating profiles in the atmosphere. Aerosol absorption optical depth (AAOD) is an important parameter for evaluating the model performance in simulating light absorbing aerosols. Figure 9 shows the seasonal mean AAOD at 500 nm averaged for 2010-2014 and over the three sub-regions from the OMI retrievals and the WRF-Chem simulation. Both retrievals and simulation show small interannual variability (not shown). Both the retrievals and simulation show 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. The simulated AAOD over West Pacific agrees reasonably well with the OMI retrieval in DJF but is higher in the other three seasons. Over Central Pacific, the simulation generally

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

reproduces the retrieved AAOD, except overestimating (underestimating) the retrieved values in JJA (DJF). Over East Pacific, the simulated AAOD is consistent with the retrieved values with some overestimation in JJA. 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 West Pacific than Central and East Pacific, which is consistent with the AOD pattern.

The AERONET retrieval products (version 2) also provide AAOD values but only at the sites and time when the total AOD exceeds a threshold value of 0.4 at 440 nm because the AERONET inversion algorithms require a high signal-to-noise ratio to retrieve some optical products such as AAOD. The total AOD values over Central Pacific and the western US are less than this threshold value most of the time, and only AAOD values retrieved at the East Asian sites are available and reliable. Figure 10 shows the monthly variation of AAOD averaged at the AERONET sites over East Asia (Fig. 1) from the AERONET observations, OMI retrievals, and WRF-Chem simulation. The AERONET retrievals show the monthly variation of AAOD over East Asia with relatively lower values in JJA probably due to wet removal of aerosols by precipitation and mixing with clean marine air during the East Asian summer monsoon (Zhao et al., 2010). The simulation generally captures the observed monthly variability, but overestimate AAOD in the warm months such as August and September. The model shows that AAOD over East Asia is dominated by BC and is partly contributed by dust. Other aerosols contribute to small amount of AAOD due to the internal mixing of aerosols in the atmosphere (Zhao et al., 2013a). The model positive biases in AAOD in the warm months may be partly related to the constant anthropogenic BC emissions

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

applied throughout the seasons while previous studies showed that anthropogenic BC emissions may have seasonal variation with lower values in the warm months (Lu et al., 2010).

It is noteworthy that the OMI retrieved AAOD is lower than that from AERONET and WRF-Chem, particularly in JJA and SON. The lower OMI AAOD may indicate positive biases in the OMI SSA retrievals over East Asia and West Pacific (Fig. 9) that is significantly affected by the East Asian outflow. The difference between the simulation and OMI retrievals over East Pacific may be partly due to negative biases in modeling SSA; however, it is noteworthy that 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 less-sufficient amounts of absorbing aerosols [Veihelmann et al., 2007; Torres et al., 2013]. In addition, the OMI retrievals may also have cloud contamination issue because of the large footprint.

4.4 Aerosol vertical distributions

Column integrated properties of aerosol (e.g., AOD and AAOD) provide useful information in regard to atmospheric aerosol loading but little information on the vertical distribution of aerosols. Previous studies found that simulated aerosol vertical distributions differ significantly, which can affect the assessments of aerosol impacts on climate and air quality (e.g., Schulz et al., 2006; Textor et al., 2006). CALIPSO with the unique capability provides an opportunity to assess model simulation of aerosol vertical distributions (e.g., Huang et al., 2013). Figure 11 shows the vertical distributions of annual mean aerosol extinction coefficients for 2010-2014 averaged over the three sub-

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

regions from the CALIPSO retrievals and the corresponding WRF-Chem simulation under cloud-free condition. The CALIPSO retrievals show clearly that aerosol extinction coefficients peak near several hundred meters above the surface and then decrease with the altitude over the three sub-regions. The extinction coefficients reduce from West to East Pacific. The model generally reproduces the aerosol extinction vertical distributions. The simulated mass fraction of each aerosol component (Fig. 12) show that below 1 km, sea-salt dominates the total aerosol mass over Central and East Pacific, while the outflow of anthropogenic aerosols and dust also make significant contributions over West Pacific. Above 4 km, dust is the dominant aerosol over all three sub-regions. The simulation is a little higher than the retrievals in the free troposphere (e.g., >4 km) and at the surface. In the free troposphere, the difference may be due to the reduced sensitivity of CALIOP to tenuous aerosol layers above 4 km. At the surface, the lower CALIPSO aerosol extinction may result from a misclassification of polluted continental aerosol as marine aerosol when pollution outflow occurs near the surface and surface contamination during the retrievals (Yu et al., 2010). The model consistently underestimates the aerosol extinction coefficients between surface and 1 km in all three sub-regions, which may indicate that the model has negative biases for estimating marine aerosol emissions, as shown in Fig. 4.

The seasonal variation of aerosol extinction profiles averaged for 2010-2014 (Fig. 13) shows the spring maximum, particularly above 2 km, over all three sub-regions from both the CALIPSO retrievals and the model simulation. This is likely due to the seasonality of dust outflow over the Pacific (Fig. 14) that dominates the aerosol masses above 2 km with a peak in spring (e.g., Huang et al. 2013). Over West Pacific, the model reasonably reproduces the retrieved aerosol extinction profiles through the seasons with a

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

relatively larger negative bias below 1 km in DJF, when sea-salt has a relatively larger contribution near the surface (Fig. 14). In JJA, similar amount of aerosols as that in MAM has the largest contribution from the anthropogenic pollutant outflow among the seasons with a peak at ~ 2 km above the surface. Over Central and East Pacific, the model also well captures the vertical distributions with a relatively larger negative bias below 1 km in DJF. Over these two regions, the seasonality of the vertical shape of each aerosol component contribution is similar to that over West Pacific, except that the sea-salt contribution is larger near the surface (Fig. 14).

4.5 Aerosol surface mass concentrations over the West US

For lack of in-situ observations of aerosol masses over the Pacific, measurements of surface fine aerosol ($PM_{2.5}$) component mass concentrations from the IMPROVE network over the western U.S. were widely used for model evaluation of trans-Pacific transport (e.g., Chin et al., 2007; Hadley et al., 2007). Daily variation of surface fine aerosols (dust, sulfate, nitrate, BC, and OC) averaged for 2010-2014 from the IMPROVE measurements and the corresponding model simulations are illustrated in Figure 15. The IMPROVE sites over the western U.S. (Fig. 1) that have measurements for the entire five years (2010-2014) and with less noisy values are divided at 40°N into two groups to represent the Northwest and Southwest U.S. The averaged values over the Northwest and Southwest sites are shown. At the Northwest sites, the model well captures the observed seasonal variation of dust. Both the observations and simulation show the maximum dust mass concentration in MAM. The observed surface sulfate concentrations are the lowest in the cold season when photochemistry is least active, and the highest in the warm

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

season when the most active photochemistry occurs. This seasonality of sulfate may also be contributed by the seasonality of wet removal (much more precipitation in DJF). Nitrate shows a seasonality that is opposite to that of sulfate, with a maximum surface concentration occurring in the cold season and a minimum in the warm season, which can be explained by the combined effects of temperature and vertical turbulent mixing (Zhao et al., 2013a). The simulation generally reproduces the magnitude and seasonality of both sulfate and nitrate. At the Southwest sites, the simulation is consistent with the observations on the magnitude and seasonality of surface concentrations of dust, surface, and nitrate.

A sensitivity simulation without dust, fire, and anthropogenic emissions over North America (10°N-70°N and 170°W-60°W) indicates that the trans-Pacific transported dust dominates the total dust amount in all seasons at both the northern and southern sites, particularly in MAM. At the southern sites, the North American dust makes a significant contribution in DJF. The sensitivity simulation also shows that trans-Pacific transported sulfate and nitrate can make significant contribution to their surface concentration over the western U.S., and the relative contributions are larger when the surface concentrations are lower (i.e., cold seasons for sulfate and warm seasons for nitrate). During the cold seasons, the North American anthropogenic emissions determine the nitrate surface concentrations. Some differences in dust, sulfate, and nitrate surface concentrations also exist between the observations and simulation. These differences may reflect partly the modeling biases of trans-Pacific aerosols, and also the uncertainties in the North American dust and anthropogenic emissions. Another source of the difference may be from the sub-grid variability of emissions and surface concentrations that

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

confounds the comparison of model simulation at one-degree horizontal grid resolution and the point measurements from the individual sites.

There is a significant difference in BC and OC surface concentrations between the observations and simulation. At the Northwest sites, the observed BC and OC show significant seasonal variation with the highest surface concentration in June-September (JJAS). This consistent peak of BC and OC surface concentrations is likely due to the North American biomass burning that also reaches a maximum in JJAS (Chin et al., 2007). The simulation captures this seasonality to some extent but significantly underestimates the JJAS peak for both BC and OC. The sensitivity simulation shows that the peak is dominated by the North American emissions. 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.

At the Southwest sites, the impact of biomass burning on the BC and OC surface concentrations seems relatively small. The simulation can well capture the magnitude and seasonality of surface BC concentration that shows the maximum in DJF and the minimum in JJA, which is likely due to stronger vertical turbulent mixing in JJA compared with DJF. The observed OC still shows peak concentrations in JJA, and the model significantly underestimates the peak OC concentrations. However, this negative bias 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 of more active photochemistry and higher emissions of biogenic VOCs. The underestimation of secondary organic aerosol (SOA)

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

may be due to uncertainty of biogenic emissions (Zhao et al., 2015) and the outdated SOA mechanism used in the current version of WRF-Chem (Shrivastava et al., 2011). On the other hand, it is also noteworthy that uncertainties in the IMPROVE carbonaceous aerosol data are also relatively high because they are inferred from optical/thermal measurements. The sensitivity simulation again shows that the peaks of BC and OC surface concentrations are dominated by the North American emissions.

5 Summary and conclusion

A fully coupled meteorology-chemistry model (WRF-Chem) has been configured to conduct quasi-global simulation for the 5 years of 2010-2014. The simulation results are evaluated for first time with various reanalysis and observational datasets, including precipitation from GPCP, wind fields from MERRA, AOD, EAE, and AAOD from MODIS, MISR, OMI, and AERONET, aerosol extinction profiles from CALIPSO, and aerosol surface mass concentrations from IMPROVE. In this study, the evaluation and analysis focus on the trans-Pacific transport region for the purpose of demonstrating the capability of using the quasi-global WRF-Chem simulation to provide consistent lateral chemical boundaries for nested regional WRF-Chem simulations that can be used to investigate the impact of trans-Pacific transported aerosols on the regional air quality and climate over the western US. The main conclusion is summarized below:

- The comparison of simulated AOD with the satellite and AERONET retrievals reveals that the model can well capture the spatial gradient of aerosol mass loading decreasing from West to East Pacific, resulting from the sea-salt loading and the Asian pollutant outflow. The seasonal variation of aerosols across Pacific with the

© Author(s) 2016. CC-BY 3.0 License.

maximum AOD in MAM is also reproduced by the model. The model underestimates AOD over the ocean to the south of 20°N and over the continent of North America against the satellite retrievals. This discrepancy may reflect the model underestimation of marine emissions and/or overestimation of aerosol wet removal or the positive retrieval errors due to cloud-contamination. Compared with the AERONET retrievals, the difference of AOD over the western US between the simulation and satellite retrievals may be due to uncertainty in the satellite retrievals over the continent.

- The assessment of simulated EAE indicates that the model captures the observed smaller-size aerosols over West Pacific contributed by the Asian pollutant outflow compared to the relatively larger particles over Central and East Pacific with more contributions from sea-salt. The model also simulates the consistent seasonality of EAE with observations showing a minimum in DJF and a maximum in JJA due to the active production of small particles in warm seasons.
- The model reasonably simulates the decreasing gradient of OMI derived AAOD from East to West of Pacific, and the seasonality with a peak in MAM due to the strong outflow of dust and anthropogenic pollutants. The comparison with AERONET retrieved AAOD over East Asia may indicate that the OMI SSA retrievals have positive biases over East Asia and also West Pacific that is significantly affected by the East Asian outflow, particularly in JJA. Over East Asia, the model positive biases in AAOD in the warm months may be partly due to the neglect of the seasonal variability of anthropogenic BC emissions in this study.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

- The model generally captures the CALIPSO retrieved vertical distributions of aerosol extinction coefficients roughly decreasing with the altitude over the Pacific. Near the surface, retrieval uncertainties and model biases in estimating marine aerosol emissions may both contribute to the discrepancy between the simulation and retrievals. The difference between the simulation and retrievals in the free troposphere may be due to the reduced sensitivity of CALIOP to the aerosol layers above 4 km. The model well captures the seasonality of aerosol extinction profiles with a maximum in MAM, which is largely controlled by the activity of dust outflow events over the Pacific.
- Compared with the measurements from the IMPROVE sites over the western US, the model reproduces the observed magnitude and seasonality of dust, sulfate, and nitrate surface concentrations, with peaks in MAM, JJA, and DJF, respectively. Over the southwestern U.S., the simulation reproduces the magnitude and seasonality of surface BC concentrations that show the maximum in DJF, but significant underestimates the surface OC concentrations in JJA likely due to the negative biases in SOA production. Over the northwestern U.S., the simulation significantly underestimates surface BC and OC concentrations likely due to the uncertainties in fire emissions of which the monthly variation and the half-degree horizontal resolution may not be sufficient to capture the strong local fire events. Another source of the difference may be due to the discrepancy in spatial scales between site observations and model output for grid cell area of one-degree resolution. In addition, uncertainties in IMPROVE may also contribute to the discrepancy, in particular for carbonaceous aerosols that are inferred from optical/thermal measurements.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

- The sensitivity simulation shows that the trans-Pacific transported dust dominates the dust surface concentrations in the western US, particularly in MAM. The trans-Pacific transported sulfate and nitrate can also make significant contribution to their surface concentration over the rural areas of the western US, in particular when their surface concentrations are relatively low. The peaks of BC and OC surface concentrations over the western US are dominated by the North American emissions. These sensitivity simulation results may be different to some extent from other models (e.g., Chin et al., 2007), which could result from the considerable differences in aerosol composition and vertical distributions due to differences in model treatments of emissions and removal processes as revealed by several inter-comparison studies (Barrie et al., 2001; Penner et al., 2002; Textor et al., 2006). More detailed model inter-comparison of the trans-Pacific transport of aerosols deserves further study.

Although dust and biomass burning emissions in general have considerable year-to-year variations, the interannual variability of seasonal AOD for 2010-2014 averaged over the three sub-regions of Pacific is small as indicated by the retrievals and simulation. It is noteworthy that the trans-Pacific aerosols identified in this study include not only the outflow of Asian pollutants and dust but also European pollutants and African dust that are transported to Asia and then merged with the Asian outflow. This has been recognized by previous studies (e.g., Chin et al., 2007) and also our on-going research (Hu et al., 2015). The evaluation in this study successfully demonstrates that the WRF-Chem quasi-global simulation can be used for studying trans-Pacific transport of aerosols and providing reasonable inflow chemical boundaries for the western U.S. to further

© Author(s) 2016. CC-BY 3.0 License.

understand the impact of transported pollutants on the air quality and regional climate with high resolution nested regional modeling. It needs to be noted that the aerosol optical properties, such as AOD, AAOD, and EAE, derived from the retrievals and simulations have some different assumptions of the physical and optical parameters, so that the link between the model and the satellite data are only qualitative or semi-quantitative. Evaluation of model results with in-situ observations, particularly for a specific event may also be needed, especially over Asia and Pacific, where data are sparse or inaccessible.

Code availability

The WRF-Chem version 3.5.1 release can be obtained at http://www2.mmm.ucar.edu/wrf/users/download/get_source.html. A general WRF-Chem user's guide is also available online (http://ruc.noaa.gov/wrf/WG11/). Code modifications and model configuration for conducting quasi-global WRF-Chem simulations here are available upon request by contacting the corresponding author and will be incorporated in the future available release of WRF-Chem.

Acknowledgements

This research was supported by the Office of Science of the U.S. Department of Energy (DOE) as part of the Regional & Global Climate Modeling (RGCM) program. J. Huang acknowledges support from the National Basic Research Program of China (2012CB955301). H. Yu was supported by NASA CALIPSO project (NNX14AB21G) managed by Dr. David Considine. This study used computing resources from the PNNL

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

Institutional Computing. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute for the DOE under contract DE-AC05-76RL01830. The CALIPSO data were obtained from the NASA Langley Research Center Atmospheric Sciences Data Center. MODIS and MISR data were obtained from the NASA Atmospheric Science Data Center. OMI data were obtained from the NASA Goddard Earth Sciences Data and Information Services Center.

© Author(s) 2016. CC-BY 3.0 License.

Reference

Ahn, C., O. Torres, and P. K. Bhartia: Comparison of Ozone Monitoring Instrument UV Aerosol Products with Aqua/Moderate Resolution Imaging Spectroradiometer and Multiangle Imaging Spectroradiometer observations in 2006, 113, D16S27, doi:10.1029/2007JD008832, 2008.

Anderson, T. L., Y. Wu, D. A. Chu, B. Schmid, J. Redemann, and O. Dubovik: Testing the MODIS satellite retrieval of aerosol fine-mode fraction, J. Geophys. Res., 110, D18204, doi:10.1029/2005JD005978, 2005.

Angstrom, A. : On the atmospheric transmission of Sun radiation and on dust in the air, Geogr. Ann, 11, 156-166, 1929.

Ault, A. P., C. R. Williams, A. B. White, P. J. Neiman, J. M. Creamean, C. J. Gaston, F. M. Ralph, and K. A. Prather: Detection of Asian dust in California orographic precipitation, J. Geophys. Res., 116, D16205, doi:10.1029/2010JD015351, 2011.

Barnard, J. C., J. D. Fast, G. Paredes-Miranda, W. P. Arnott, and A. Laskin: Technical Note: Evaluation of the WRF-Chem "Aerosol Chemical to Aerosol Optical Properties" Module using data from the MILAGRO campaign, Atmos. Chem. Phys., 10, 7325-7340, doi:10.5194/acp-10-7325-2010, 2010.

Barrie, L. A., Y. Yi, W. R. Leaitch, U. Lohmann, P. Kasibhatla, G. J. Roelofs, J. Wilson, F. McGovern, C. Benkovitz, M. A. Melieres, K. Law, J. Prospero, M. Kritz, D. Bergmann, C. Bridgeman, M. Chin, J. Christensen, R. Easter, J. Feichter, C. Land, A. Jeuken, E. Kjellstrom, D. Koch, and P. Rasch: A comparison of large-scale atmospheric sulphate aerosol models (COSAM): overview and highlights, Tellus, 53B, 615-645, 2001.

© Author(s) 2016. CC-BY 3.0 License.

Binkowski, F. S., and Shankar, U.: The Regional Particulate Matter Model 1. Model description and preliminary results, J. Geophys. Res., 100(D12), 26191-26209, 1995.

Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, Atmos. Chem. Phys., 9, 945–964, doi:10.5194/acp-9-945-2009, 2009.

Chen, S., J. Huang, C. Zhao, Y. Qian, L. R. Leung, and B. Yang: Modeling the transport and radiative forcing of Taklimakan dust over the Tibetan Plateau: A case study in the summer of 2006, J. Geophys. Res., 118(2), 797-812, doi:10.1002/jgrd.50122, 2013.

Chen, S., C. Zhao, Y. Qian, L. R. Leung, J. Huang, Z. Huang, J. Bi, W. Zhang, J. Shi, L. Yang, D. Li, J. Li: Regional modeling of dust mass balance and radiative forcing over East Asia using WRF-Chem, Aeolian Research, 15, 15-30, doi:10.1016/j.aeolia.2014.02.001, 2014.

Chin, M., A. Chu, R. Levy, L. Remer, Y. Kaufman, B. Holben, T. Eck, P. Ginoux, and Q. Gao: Aerosol distribution in the Northern Hemisphere during ACE-Asia: Results from global model, satellite observations, and Sun photometer measurements, J. Geophys. Res., 109, D23S90, doi:10.1029/2004JD004829, 2004.

Chin, M., T. Diehl, P. Ginoux, and W. Malm: Intercontinental transport of pollution and dust aerosols: implications for regional air quality, Atmos. Chem. Phys., 7, 5501–5517, 2007.

Creamean, J. M., K. J. Suski, D. Rosenfeld, A. Cazorla, P. J. DeMott, R. C. Sullivan, A. B. White, F. M. Ralph, P. Minnis, J. M. Comstock, J. M. Tomlinson, and K. A. Prather: Dust and Biological Aerosols from the Sahara and Asia Influence

© Author(s) 2016. CC-BY 3.0 License.

Precipitation in the Western U.S., Science, 339, 1572, DOI: 10.1126/science.1227279, 2013.

Dentener, F., S. Kinne, T. Bond, O. Boucher, J. Cofala, S. Generoso, P. Ginoux, S. Gong, J. J. Hoelzemann, A. Ito, L. Marelli, J. E. Penner, J. P. Putaud, C. Textor, M. Schulz, G. R. van der Werf, and J. Wilson: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom: Atmos. Chem. Phys., 6, 4321-4344, 2006.

Diner, D. J., J. C. Beckert, T. H. Reilly, C. J. Bruegge, J. E. Conel, R. A. Kahn, J. V. Martonchik, T. P. Ackerman, R. Davies, S. A. W. Gerstl, H. R. Gordon, J. P. Muller, R. B. Myneni, P. J. Sellers, B. Pinty, and M. M. Verstraete: Multi-angle Imaging SpectroRadiometer (MISR) Instrument Description and Experiment Overview, IEEE Trans, Geosci. Remote Sens, 36, 1072-1087, 1998.

Dubovik, O. and King, M. D.: A flexible inversion algorithm for retrieval of aerosol optical properties from Sum and sky radiance measurements, J. Geophys. Res., 105(16), 20673-20696, 2000.

Dubovik, O., B. N. Holben, T. Lapyonok, A. Sinyuk, M. I. Mishchenko, P. Yang, and I. Slutsker: Non-spherical aerosol retrieval method employing light scattering by spheroids, Geophys. Res. Lett., 29(10), 1415, 10.1029/2001GL014506, 2002.

Easter, R. C., Ghan, S. J., Zhang, Y., Saylor, R. D., Chapman, E. G., Laulainen, N. S., Abdul-Razzak, H., Leung, L. R., Bian, X., and Zaveri, R. A.: MIRAGE: Model Description and Evaluation of Aerosols and Trace Gases, J. Geophys. Res., 109, D20210, doi:10.1029/2004JD004571, 2004.

© Author(s) 2016. CC-BY 3.0 License.

Eck, T. F., B. N. Holben, J. S. Reid, O. Dubovik, A. Smirnov, N. T. O'Neill, I. Slutsker, and S. Kinn: Wavelength dependence of the optical depth of biomass burning urban, and desert dust aerosols, , J. Geophys. Res., 104(24), 31333-31349, 1999.

Eguchi, K., I. Uno, K. Yumimoto, T. Takemura, A. Shimizu, N. Sugimoto, and Z. Liu: Trans-pacific dust transport: integrated analysis of NASA/CALIPSO and a global aerosol transport model, Atmos. Chem. Phys., 9, 3137–3145, 2009.

Fairlie, T. D., D. J. Jacob, and R. J. Park: The impact of transpacific transport of mineral dust in the United States: Atmospheric Environment, 41, 1251-1266, 2007.

Fan, J., L. R. Leung, P. J. DeMott, J. M. Comstock, B. Singh, D. Rosenfeld, J. M. Tomlinson, A. White, K. A. Prather, P. Minnis, J. K. Ayers, and Q. Min: Aerosol impacts on California winter clouds and precipitation during CalWater 2011: local pollution versus long-range transported dust, Atmos. Chem. Phys., 14, 81–101, doi:10.5194/acp-14-81-2014, 2014.

Fan, J., D. Rosenfeld, Y. Yang, C. Zhao, L. R. Leung, and Z. Li: Substantial contribution of anthropogenic air pollution to catastrophic floods in Southwest China, Geophys. Res. Lett., 42, 6066-6075, doi:10.1002/2015GL064479, 2015.

Fast, J. D., W. I. Gustafson, R. C. Easter, R. A. Zaveri, J. C. Barnard, E. G. Chapman, G. A. Grell, and S. E. Peckham: Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry- aerosol model, J. Geophys. Res., 111, D21305, doi:10.1029/2005JD006721, 2006.

Fast, J., A. C. Aiken, J. Allan, L. Alexander, T. Campos, M. R. Canagaratna, E. Chapman, P. F. DeCarlo, B. de Foy, J. Gaffney, J. de Gouw, J. C. Doran, L. Emmons,

© Author(s) 2016. CC-BY 3.0 License.

A. Hodzic, S. C. Herndon, G. Huey, J. T. Jayne, J. L. Jimenez, L. Kleinman, W. Kuster, N. Marley, L. Russell, C. Ochoa, T. B. Onasch, M. Pekour, C. Song, I. M. Ulbrich, C. Warneke, D. Welsh-Bon, C. Wiedinmyer, D. R. Worsnop, X. Y. Yu, and R. Zaveri: Evaluating simulated primary anthropogenic and biomass burning organic aerosols during MILAGRO: implications for assessing treatments of secondary organic aerosols, Atmos. Chem. Phys., 9, 6191–6215, 2009.

Fast, J. D., J. Allan, R. Bahreini, J. Craven, L. Emmons, R. Ferrare, P. L. Hayes, A. Hodzic, J. Holloway, C. Hostetler, J. L. Jimenez, H. Jonsson, S. Liu, Y. Liu, A. Metcalf, A. Middlebrook, J. Nowak, M. Pekour, A. Perring, L. Russell, A. Sedlacek, J. Seinfeld, A. Setyan, J. Shilling, M. Shrivastava, S. Springston, C. Song, R. Subramanian, J. W. Taylor, V. Vinoj, Q. Yang, R. A. Zaveri, and Q. Zhang: Modeling regional aerosol and aerosol precursor variability over California and its sensitivity to emissions and long-range transport during the 2010 CalNex and CARES campaigns, Atmos. Chem. Phys., 14, 10013–10060, doi:10.5194/acp-14-10013-2014, 2014.

Fischer, E. V., N. C. Hsu, D. A. Jaffe, M.-J. Jeong, and S. L. Gong: A decade of dust: Asian dust and springtime aerosol load in the U.S. Pacific Northwest, Geophys. Res. Lett., 36, L03821, doi:10.1029/2008GL036467, 2009.

Forster, C., O. Cooper, A. Stohl, S. Eckhardt, P. James, E. Dunlea, D. K. Nicks, J. S. Holloway, G. Hubler, D. D. Parrish, T. B. Ryerson, and M. Trainer: Lagrangian transport model forecasts and a transport climatology for the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) measurement campaign, J. Geophys. Res., 109, D07S92, doi:10.1029/2003JD003589, 2004.

© Author(s) 2016. CC-BY 3.0 License.

Gao, Y., X. Liu, C. Zhao, and M. Zhang: Emission controls versus meteorological conditions in determining aerosol concentrations in Beijing during the 2008 Olympic Games, Atmos. Chem. Phys., 11, 12437–12451, doi:10.5194/acp-11-12437-2011, 2011.

Gao, Y., C. Zhao, X. H. Liu, M. G. Zhang, and L. R. Leung: WRF-Chem simulations of aerosols and anthropogenic aerosol radiative forcing in East Asia, Atmospheric Environment, 92, 250-266, 2014.

Ginoux, P., M. Chin, I. Tegen, J. M. Prospero, B. Holben, O. Dubovik, and S. J. Lin: Sources and distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res., 106(17), 20255-20273, 2001.

Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global Biogeochemical Cycle, 17(4), 1097, doi:10.1029/2003GB002079, 2003.

Grell, G. A., S. E. Peckham, R. Schmitz, S. A. McKeen, G. Frost, W. C. Skamarock, and B. Eder: Fully coupled ‘‘online’’ chemistry within the WRF model, Atmospheric Enviroment, 39, 6957-6957, doi:10.1016/j.atmosenv.2005.04.027, 2005.

Gustafson, W. I., E. G. Chapman, S. J. Ghan, R. C. Easter, and J. D. Fast: Impact on modeled cloud characteristics due to simplified treatment of uniform cloud condensation nuclei during NEAQS 2004, Geophys. Res. Lett., 34, L19809, doi:10.1029/2007GL030021, 2007.

Hadley, O. L., V. Ramanathan, G. R. Carmichael, Y. Tang, C. E. Corrigan, G. C. Roberts, and G. S. Mauger: Trans-Pacific transport of black carbon and fine aerosols

© Author(s) 2016. CC-BY 3.0 License.

(D < 2.5 mm) into North America, J. Geophys. Res., 112, D05309, doi:10.1029/2006JD007632, 2007.

Hadley, O. L., C. E. Corrigan, T. W. Kirchstetter, S. S. Cliff, and V. Ramanathan: Measured black carbon deposition on the Sierra Nevada snow pack and implication for snow pack retreat, Atmos. Chem. Phys., 10, 7505-7513, doi:10.5194/acp-10-7505-2010, 2010.

Hand, J., S. A. Copeland, D. E. Day, A. M. Dillner, H. Indresand, W. C. Malm, C. E. McDade, C. T. Moore, Jr, M. L. Pitchford, B. A. Schichtel, and J. G. Watson: Spatial and seasonal patterns and temporal variability of haze and its constituents in the United States: Report V, June, 2011, available from http://vista.cira.colostate.edu/improve/Publications/ Reports/2011/2011.htm, 2011.

Heald, C. L., D. J. Jacob, R. J. Park, B. Alexander, T. D. Fairlie, R. M. Yantosca, and D. A. Chu: Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States, J. Geophys. Res., 111, D14310, doi:10.1029/2005JD006847, 2006.

Hess, M., P. Koepke, and I. Schult: Optical Properties of Aerosols and Clouds: The Software Package OPAC, Bull. Am. Meteorol. Soc., 79(5), 831-844, 1998.

Holben, B. N., T. F. Eck, I. Slutsker, D. Tanre´, J. P. Buis, A. Setzer, E. Vermote, J. A. Reagan, Y. J. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov: AERONET - A Federated Instrument Network and Data Archive for Aerosol Characterization, Remote Sens. Environ., 66, 1-16, 1998.

Holben, B. N., D. Tanr, A. Smirnov, T. F. Eck, I. Slutsker, N. Abuhassan, W. W. Newcomb, J. S. Schafer, B. Chatenet, F. Lavenu, Y. J.Kaufman, J. V. Castle, A.

© Author(s) 2016. CC-BY 3.0 License.

Setzer, B. Markham, D. Clark, R. Frouin, R. Halthore, A. Karneli, N. T. O'Neill, C. Pietras, R. T. Pinker, K. Voss, and G. Zibordi: An emergingground-basedaerosolclimatology: Aerosol optical depth from AERONET, J. Geophys. Res., 106(11), 12067-12097, 2001.

Hsu, N. C., S. C. Tsay, M. D. King, and J. R. Herman: Deep Blue Retrievals of Asian Aerosol Properties During ACE-Asia, IEEE Trans, Geosci. Remote Sens., 44(11), 3180-3195, 2006.

Hsu, N. C., M.-J. Jeong, and C. Bettenhausen: Enhanced Deep Blue aerosol retrieval algorithm: The second generation, J. Geophys. Res. Atmos., 118 (16): 9296–9315, 2013.

Hu, Z., C. Zhao, J. Huang, L. R. Leung, and Y. Qian: Transpacific transport and evolution of aerosols: Source contribution analysis, manuscript in preparation.

Huang, L., J. H. Jiang, J. L. Tackett, H. Su, and R. Fu: Seasonal and diurnal variations of aerosol extinction profile and type distribution from CALIPSO 5-year observations, J. Geophys. Res. Atmos., 118, 4572–4596, doi:10.1002/jgrd.50407, 2013.

Huffman, G. J., R. F. Adler, M. M. Morrissey, D. T. Bolvin, S. Curtis, R. Joyce, B. McGavock, and J. Susskind: Global Precipitation at One-Degree Daily Resolution from Multisatellite Observations. J. Hydrometeor, 2, 36–50,doi: http://dx.doi.org/10.1175/1525-7541(2001)002<0036:GPAODD>2.0.CO;2, 2001.

Iacono, M. J., E. J. Mlawer, S. A. Clough, and J. J. Morcrette: Impact of an improvedlongwaveradiation model,RRTM, on the energy budget and thermodynamic properties of the NCAR community climate model, CCM3, J. Geophys. Res., 105(11), 14873-14890, 2000.

© Author(s) 2016. CC-BY 3.0 License.

Jaegle, L., P. K. Quinn, T. S. Bates, B. Alexander, and J. T. Lin: Global distribution of sea salt aerosols: new constraints from in situ and remote sensing observations, Atmos. Chem. Phys., 11, 3137-3157, doi:10.5194/acp-11-3137-2011, 2011.

Jaffe, D., T. Anderson, D. Covert, R. Kotchenruther, B. Trost, J. Danielson, W. Simpson, T. Berntsen, S. Karlsdottir, D. Blake, J. Harris, G. Carmichael, and I. Uno: Transport of Asian Air Pollution to North America, Geophys. Res. Lett., 26(6), 711-714, 1999.

Jethva, H., O, Torres, and C. Ahn: Global assessment of OMI aerosol single-scattering albedo using ground-based AERONET inversion, J. Geophys. Res. Atmos., 119, 9020–9040, doi:10.1002/ 2014JD021672, 2014.

Kahn, R. A., D. L. Nelson, M. J. Garay, R. C. Levy, M. A. Bull, D. J. Diner, J. V. Martonchik, S. R. Paradise, E. G. Hansen, and L. A. Remer: MISR Aerosol Product Attributes and Statistical Comparisons With MODIS, IEEE Trans, Geosci. Remote Sens, 47(12), 4095-4114, 2009.

Kaufman, Y. J., D. Tanré, L. A. Remer, E. F. Vermote, A. Chu, and B. N. Holben: Operational remote sensing of tropospheric aerosol over land from EOS moderate resolution imaging spectroradiometer, J. Geophys. Res., 102(D14), 17051-17067, 1997.

Kaufman, Y. J., O. Boucher,D. Tanre, M. Chin, L. A. Remer, and T. Takemura: Aerosol anthropogenic component estimated from satellite data, Geophys. Res. Lett., 32, L17804, doi:10.1029/2005GL023125, 2005a.

Kaufman, Y. J., I. Koren, L. A. Remer, D. Rosenfeld, and Y. Rudich: The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic

© Author(s) 2016. CC-BY 3.0 License.

Ocean, Proc. Natl. Acad. Sci. USA, 102(32), 11207-11212, Doi/10.1073/pnas.0505191102, 2005b.

King, M. D., Y. J. Kaufman, D. Tanre, and T. Nakajima: Remote Sensing of Tropospheric Aerosols from Space: Past, Present, and Future, Bull. Am. Meteorol. Soc., 80(11), 2229-2259, 1999.

Kok, J. F.: A scaling theory for the size distribution of emitted dust aerosols suggests climate models underestimate the size of the global dust cycle, Proc. Natl. Acad. Sci. USA, 108(3), 1016-1021, doi/10.1073/pnas.1014798108, 2011.

Koren, I., Y. J. Kaufman, D. Rosenfeld, L. A. Remer, and Y. Rudich: Aerosol invigoration and restructuring of Atlantic convective clouds, Geophys. Res. Lett., 2, L14828, doi:10.1029/2005GL023187, 2005

Levy, R. C., L. A. Remer, R. G. Keidman, S. Mattoo, C. Ichoku, R. Kahn, and T. F. Eck: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, Atmos. Chem. Phys., 10, 10399–10420, doi:10.5194/acp-10-10399-2010, 2010.

Levy, R. C., S. Mattoo, L. A. Munchak, L. A. Remer, A. M. Sayer, F. Patadia, and N. Hsu: The Collection 6 MODIS Aerosol Products over Land and Ocean, Atmos. Meas. Tech., 6, 2989-3034 doi:10.5194/amt-6-2989-2013, 2013.

Liang, Q., L. Jaegle, D. A. Jaffe, P. Weiss-Penzias, A. Heckman, and J. A. Snow: Long-range transport of Asian pollution to the northeast Pacific: Seasonal variations and transport pathways of carbon monoxide, J. Geophys. Res., 109, D23S07, doi:10.1029/2003JD004402, 2004.

Liu, Z., M. A. Vaughan, D. M. Winker, C. A. Hostetler, L. R. Poole, D. Hlavka, W. Hart, abd M. McGill: Use of probability distribution functions for discriminating between

© Author(s) 2016. CC-BY 3.0 License.

cloud and aerosol in lidar backscatter data, J. Geophys. Res., 109, D15202, doi:10.1029/2004JD004732, 2004.

Lu, Z., D. G. Streets, Q. Zhang, S. Wang, G. R. Carmichael, Y. F. Cheng, C. Wei, M. Chin, T. Diehl, and Q. Tan: Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000, Atmos. Chem. Phys., 10, 6311-6331, doi:10.5194/acp-10-6311-2010, 2010.

Lu, Z., Q. Zhang, and D. G. Streets: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, Atmos. Chem. Phys., 11, 9839-9864, doi:10.5194/acp-11-9839-2011, 2011.

Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill: Spatial and seasonal trends in particle concentration and optical extinction in the United States, 99(D1), 1347-1370, 1994.

Martonchik, J. V., David J. D., Kathleen A. C., and M. A. Bull: Regional Aerosol Retrieval Results From MISR, IEEE Trans, Geosci. Remote Sens, 40(7), 1520-1531, 2002.

McKeen, S. A., G. Wotawa, D. D. Parrish, J. S. Holloway, M. P. Buhr, G. Hubler, F. C. Fehsenfeld, and J. F. Meagher: Ozone production from Canadian wildfires during June and July of 1995, J. Geophys. Res., 107(D14), 4192, 10.1029/2001JD000697, 2002.

Mlawer, E. J., S. J. Taubman, P. D. Brown, M. J. Iacono, and S. A. Clough: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, J. Geophys. Res., 102(D14), 16663-16682, 1997.

© Author(s) 2016. CC-BY 3.0 License.

Painter, T. H., J. S. Deems, J. Belnap, A. F. Hamlet, C. C. Landry, and B. Udall: Response of Colorado River runoff to dust radiative forcing in snow, Proc. Natl. Acad. Sci. USA, 107(40), 17125-17130, doi/10.1073/pnas.0913139107, 2010.

Penner, J. E., S. Y. Zhang, M. Chin, C. C. Chuang, J. Fechter, Y. Feng, I. V. Geogdzhayev, P. Ginoux, M. Herzog, A. Higurashi, D. Koch, C. Land, U. Lohmann, M. Mischecko, T. Nakajima, G. Pitari, B. Soden, I. Tegen, and L. Stowe: A Comparison of Model- and Satellite-Derived Aerosol Optical Depth and Reflectivity, J. Atmos. Sci., 59, 441-460, 2002.

Qian, Y., W. I. Gustafson, L. R. Leung, and S. J. Ghan: Effects of soot-induced snow albedo change on snowpack and hydrological cycle in western United States based on Weather Research and Forecasting chemistry and regional climate simulations, J. Geophys. Res., 114, D03108, doi:10.1029/2008JD011039, 2009.

Qian, Y., W. I. Gustafson Jr. and J. D. Fast: An investigation of the sub-grid variability of trace gases and aerosols for global climate modeling, Atmos. Chem. Phys., 10, 6917-6946, doi:10.5194/acp-10-6917-2010, 2010.

Qian, Y., H. Yan, Z. Hou, C. Johannesson, S. Klein, D. Lucas, R. Neale, P. Rasch, L. Swiler, J. Tannahill, H. Wang, M. Wang, and C. Zhao: Parametric sensitivity analysis of precipitation at global and local scales in the Community Atmosphere Model CAM5, J. Adv. Model. Earth Syst., 7, 382-411, doi:10.1002/2014MS000354, 2015.

Remer, L. A., Y. J. Kaufman, D. Tanre, S. Mattoo, D. A. Chu, J. V. Martins, R. R. Li, C. Ichoku, R. C. Levy, R. G. Kleidman, T. F. Eck, E. Vermote, and B. N. Holben: The MODIS aerosol algorithm, products and validation, J. Atmos. Sci., 62, 947-973, 2005.

© Author(s) 2016. CC-BY 3.0 License.

Remer, L. A. and Kaufman, Y. J.: Aerosol direct radiative effect at the top of the atmosphere over cloud free ocean derived from four years of MODIS data, Atmos. Chem. Phys., 6, 237–253, 2006.

Rienecker, M. M., M. J. Suarez, R. Gelaro, R. Todling, J. Bacmeister, E. Liu, M. G. Bosilovich, S. D. Schubert, L. Takacs, G.-K. Kim, S. Bloom, J. Chen, D. Collins, A. Conaty, A. da Silva, W. Gu, J. Joiner, R. D. Koster, R. Lucchesi, A. Molod, T. Owens, S. Pawson, P. Pegion, C. R. Redder, R. Reichle, F. R. Robertson, A. G. Ruddick, M. Sienkiewicz, and J. Woollen: MERRA: NASA's Modern-Era Retrospective Analysis for Research and Applications. J. Climate, 24, 3624–3648. doi: http://dx.doi.org/10.1175/JCLI-D-11-00015.1, 2011.

Sassen, K.: Indirect climate forcing over the western US from Asian Dust strom, Geophys. Res. Lett., 29(10), 1465, 10.1029/2011GL014051, 2002.

Schulz, M., C. Textor, S. Kinne, Y. Balkanski, S. Bauer, T. Berntsen, T. Berglen, O. Boucher, F. Dentener, S. Guibert, I. S. A. Isaksen, T. Iversen, D. Koch, A. Kirkevag, X. Liu, V. Montanaro, G. Myhre, J. E. Penner, G. Pitari, S. Reddy, Ø. Seland, P. Stier, and T. Takemura: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations, Atmos. Chem. Phys., 6, 5225–5246, 2006.

Schuster, G. L., O. Dubovik, and B. N. Holben: Angstrom exponent and bimodal aerosol size distributions, J. Geophys. Res., 111, D07207, doi:10.1029/2005JD006328, 2006.

Shrivastava, M., J. Fast, R. Easter, W. I. Gustafson, R. A. Zaveri, J. L. Jimenez, P. Saide, and A. Hodzic: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, 11, 6639–6662, doi:10.5194/acp-11-6639-2011, 2011.

© Author(s) 2016. CC-BY 3.0 License.

Skamarock, W. C. and Klemp, J. B.: A time-split nonhydrostatic atmospheric model for weather research and forecasting applications, J. Com. Phy., 227, 3456-3485, doi:10.1016/j.jcp.2007.01.037, 2008.

Stauffer, D. R. and Seaman, N. L.: Use of Four-Dimensional Data Assimilation in a Limited_Area Mesoscale Model. Part I: Experiments with Synoptic-Scale Data, Mon. Wea. Rev., 118, 1250-1277, 1990.

Tao, Z., H. Yu, and M. Chin, Impact of transpacific aerosol on air quality over the United States: A perspective from aerosol-cloud-radiation interactions, Atmos. Environ., 125, 48-60, 2016.

Textor, C., M. Schulz, S. Guibert, S. Kinne, Y. Balkanski, S. Bauer, T. Berntsen, T. Berglen, O. Boucher, M. Chin, F. Dentener, T. Diehl, R. Easter, H. Feichter, D. Fillmore, S. Ghan, P. Ginoux, S. Gong, A. Grini, J. Hendricks, L. Horowitz, P. Huang, I. Isaksen, T. Iversen, S. Kloster, D. Koch, A. Kirkevag, J. E. Kristjansson, M. Krol, A. Lauer, J. F. Lamarque, X. Liu, V. Montanaro, G. Myhre, J. Penner, G. Pitari, S. Reddy, Ø. Seland, P. Stier, T. Takemura, and X. Tie: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777–1813, 2006.

Torres, O., C. Ahn, and Z. Chen: Improvements to the OMI near-UV aerosol algorithm using A-train CALIOP and AIRS observations, Atmos. Meas. Tech., 6, 3257–3270, doi:10.5194/amt-6-3257-2013, 2013.

VanCuren, R. A.: Asian aerosols in North America: Extracting the chemical composition and mass concentration of the Asian continental aerosol plume from long-term

© Author(s) 2016. CC-BY 3.0 License.

aerosol records in the western United States, J. Geophys. Res., 108, NO. D20, 4623, doi:10.1029/2003JD003459, 2003.

van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, M. Mu, P. S. Kasibhatla, D. C. Morton, R. S. DeFries, Y. Jin, and T. T. van Leeuwen: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707–11735, doi:10.5194/acp-10-11707-2010, 2010.

Veihelmann, B., P. F. Levelt, P. Stammes, and J. P. Veefkind: Simulation study of the aerosol information content in OMI spectral reflectance measurements, Atmos. Chem. Phys., 7, 3115–3127, 2007.

Winker, D. M., W. H. Hunt, and M. J. McGill: Initial performance assessment of CALIOP, J. Geophys. Res., 34, L19803, doi:10.1029/2007GL030135, 2007.

Winker, D. M., M. A. Vaughan, A. Omar, Y. Hu, K. A. Powell, Z. Liu, W. H. Hunt, and S. A. Young: Overview of the CALIPSO Mission and CALIOP Data Processing Algorithms, J. Atmos. Oceanic Technol., 26, 2310–2323, doi: http://dx.doi.org/10.1175/2009JTECHA1281.1, 2009.

Yu, H. B., R. E. Dickinson, M. Chin, Y. J. Kaufman, B. N. Holben, I. V. Geogdzhayev, and M. I. Mishchenko: Annual cycle of global distributions of aerosol optical depth from integration of MODIS retrievals and GOCART model simulations, J. Geophys. Res.,108, NO. D3, 4128, doi:10.1029/2002JD002717, 2003.

Yu, H. B., R. E. Dickinson, M. Chin, Y. J. Kaufman, M. Zhou, L. Zhou, Y. Tian, O. Dubovik, and B. N. Holben: Direct radiative effect of aerosols as determined from a

© Author(s) 2016. CC-BY 3.0 License.

combination of MODIS retrievals and GOCART simulations, J. Geophys. Res.,109, D03206, doi:10.1029/2003JD003914, 2004.

Yu, H. B., R. Fu, R. E. Dickinson, Y. Zhang, M. Chen, and H. Wang, Interannual variability of smoke and warm cloud relationships in the Amazon as inferred from MODIS retrievals, Remote Sens. Environ., 111, 435-449, doi:10.1016/j.rse.2007.04.003, 2007.

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., M. Chin, L. A. Remer, R. G. Kleidman, N. Bellouin, H. Bian, and T. Diehl: Variability of marine aerosol fine-mode fraction and estimates of anthropogenic aerosol component over cloud-free oceans from MODIS, J. Geophys. Res., 114, D10206, doi:10.1029/2008JD010648, 2009.

Yu, H. B., M. Chin, D. M. Winker, A. H. Omar, Z. Y. Liu, C. Kittaka, and T. Diehl: Global view of aerosol vertical distributions from CALIPSO lidar measurements and GOCART simulations: Regional and seasonal variations, J. Geophys. Res.,115, D00H30, doi:10.1029/2009JD013364, 2010.

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.

Yu, H., M. Chin, H. Bian, T. L. Yuan, J. M. Prospero, A. H. Omar, L. A. Remer, D. M. Winker, Y. Yang, Y. Zhang, and Z. Zhang: Quantification of trans-Atlantic dust transport from seven-year (2007-2013) record of CALIPSO lidar measurements,

© Author(s) 2016. CC-BY 3.0 License.

Remote Sens. Environ., 159, 232-249, http://dx.doi.org/10.1016/j.rse.2014.12.010, 2015.

Zaveri, R. A. and Peters, L. K.: A new lumped structure photochemical mechanism for large-scale applications, J. Geophys. Res., 104(D23), 30387-30415, 1999.

Zaveri, R. A., R. C. Easter, J. D. Fast, and L. K. Peters: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), J. Geophys. Res., 113, D13204, doi:10.1029/2007JD008782, 2008.

Zhang, J. and Reid, J. S.: MODIS aerosol product analysis for data assimilation: Assessment of over-ocean level 2 aerosol optical thickness retrievals, J. Geophys. Res., 111, D22207, doi:10.1029/2005JD006898, 2006.

Zhang, Q., D. G. Streets, G. R. Carmichael, K. B. He, H. Huo, A. Kannari, Z. Klimont, I. S. Park, S. Reddy, J. S. Fu, D. Chen, L. Duan, Y. Lei, L. T. Wang, and Z. L. Yao: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, 2009.

Zhao, C., X. Liu, L. R. Leung, B. Johnson, S. A. McFarlane, W. I. Gustafson, J. D. Fast, and R. Easter: The spatial distribution of mineral dust and its shortwave radiative forcing over North Africa: modeling sensitivities to dust emissions and aerosol size treatments, Atmos. Chem. Phys., 10, 8821–8838, doi:10.5194/acp-10-8821-2010, 2010a.

Zhao, C., Y. Wang, Q. Yang, R. Fu, and Y. Choi: Impact of East Asia summer monsoon on the air quality over China: view from space, J. Geophys. Res., 115, D09301, doi:10.1029/2009JD012745, 2010.

© Author(s) 2016. CC-BY 3.0 License.

Zhao, C., X. Liu, L. R. Leung, and S. Hagos: Radiative impact of mineral dust on monsoon precipitation variability over West Africa, Atmos. Chem. Phys., 11, 1879-1893, doi:10.5194/acp-11-1879-2011, 2011.

Zhao, C., X. Liu, and L. R. Leung: Impact of the Desert dust on the summer monsoon system over Southwestern North America, Atmos. Chem. Phys., 12, 3717–3731, d oi:10.5194/acp-12-3717-2012, 2012.

Zhao, C., L. R. Leung, R. Easter, J. Hand, and J. Avise: Characterization of speciated aerosol direct radiative forcing over California, J. Geophys. Res., 118, 2372–2388, doi:10.1029/2012JD018364, 2013a.

Zhao, C., S. Chen, L. R. Leung, Y. Qian, J. F. Kok, R. A. Zaveri, and J. Huang: Uncertainty in modeling dust mass balance and radiative forcing from size parameterization, Atmos. Chem. Phys., 13, 10733–10753, doi:10.5194/acp-13-10733-2013, 2013b.

Zhao, C., Z. Hu, Y. Qian, L. R. Leung, J. Huang, M. Huang, J. Jin, M. G. Flanner, R. Zhang, H. Wang, H. Yan, Z. Lu, and D. G. Streets: Simulating black carbon and dust and their radiative forcing in seasonal snow: a case study over North China with field campaign measurements, Atmos. Chem. Phys., 14, 11475–11491, doi:10.5194/acp-14-11475-2014, 2014.

Zhao, C., M. Huang, J. Fast, L. Berg, Y. Qian, A. Guenther, D. Gu, M. Shrivastava, Y. Liu, S. Walters, G. Pfister, J. Jin: Sensitivity of biogenic volatile organic compounds (BVOCs) to land surface processes and vegetation distributions in California, submitted to *Geosci. Model Dev.*

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

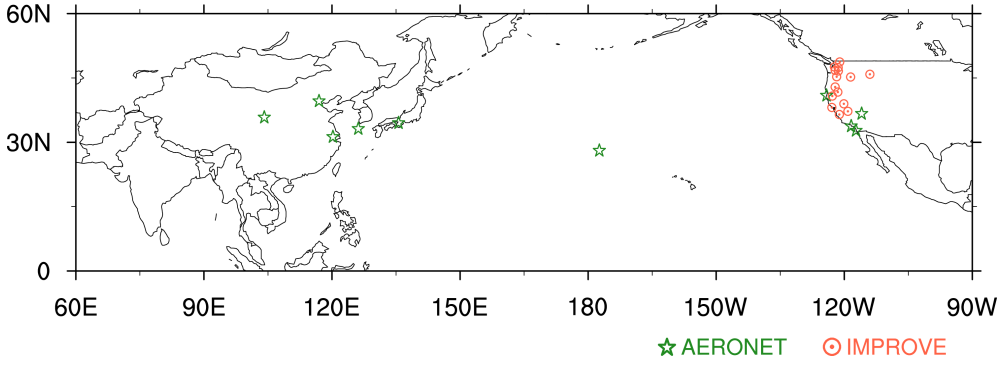

Figure 1 Observation sites for the AERONET (green stars) and IMPROVE (red dot circle) networks used in this study.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

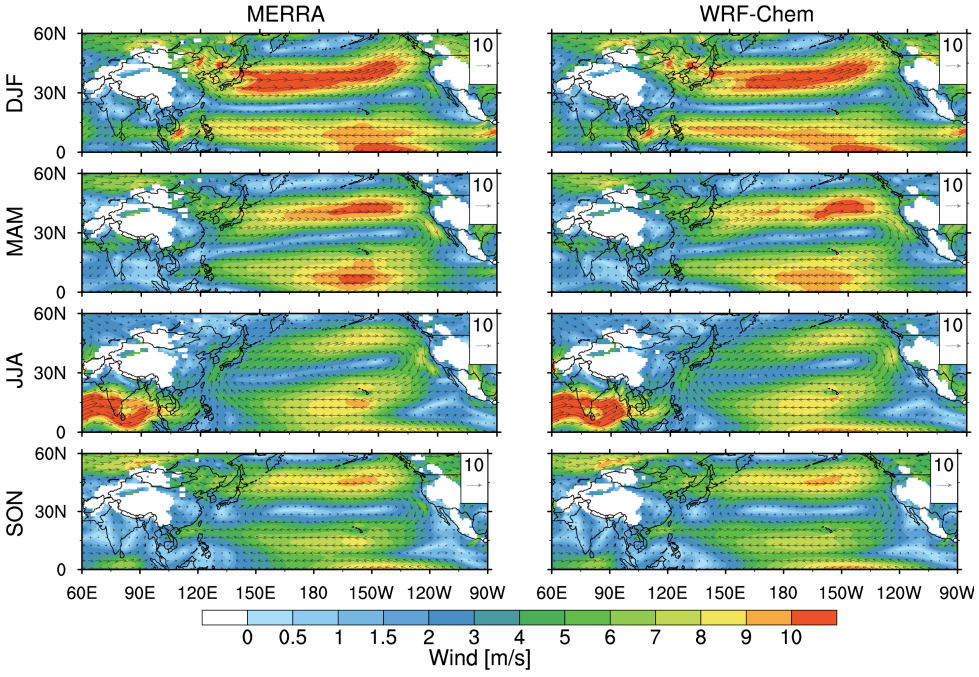

Figure 2 Spatial distributions of seasonal averaged wind fields at 850hPa from MERRA reanalysis and WRF-Chem simulations for the period of 2010-2014.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

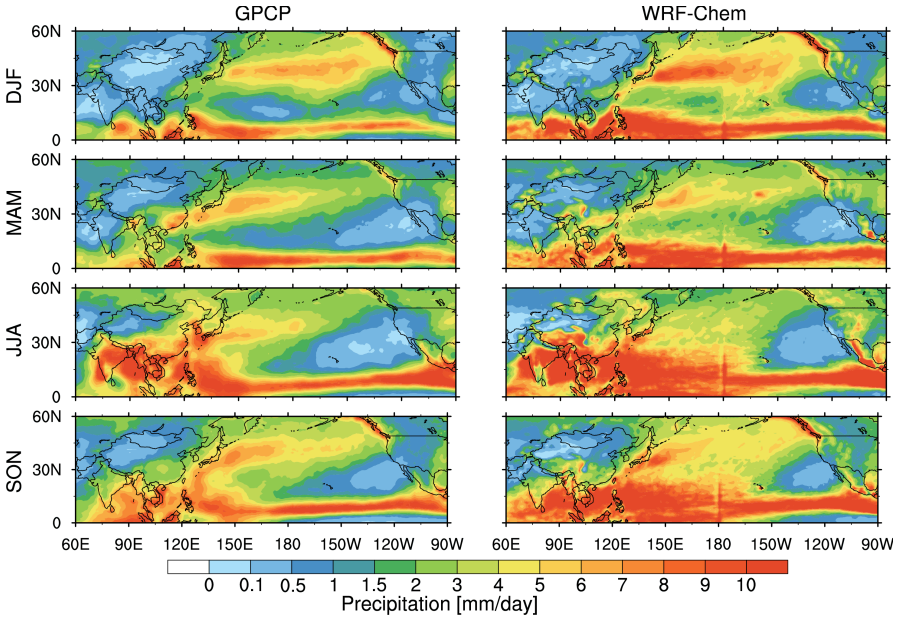

Figure 3 Spatial distributions of seasonal averaged precipitation from GPCP observations and WRF-Chem simulations for the period of 2010-2014.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

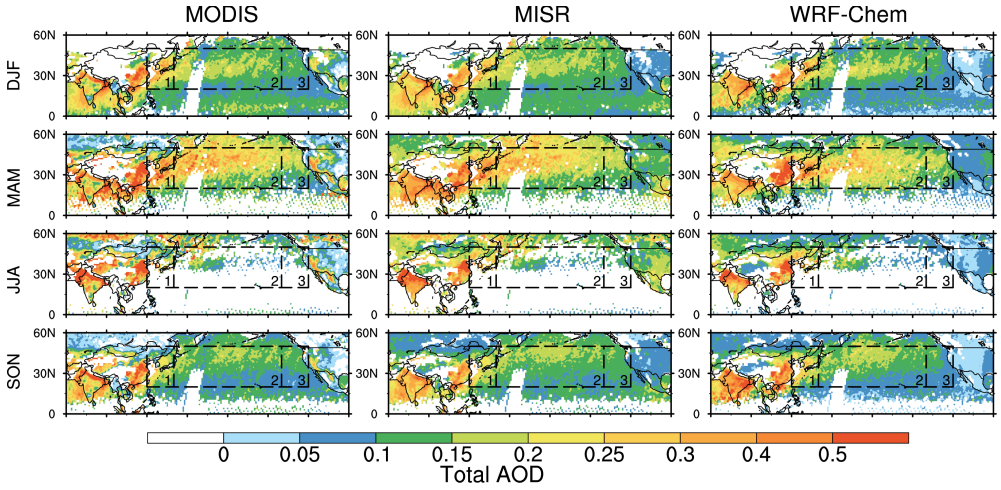

Figure 4 Spatial distributions of seasonal mean 550 nm AOD from the retrievals of MODIS and MISR onboard Terra and the WRF-Chem simulations for the period of 2010-2014. The daily results from MISR, MODIS, and WRF-Chem are only sampled for average when all of them have valid values at the same location and time. Three sub-regions are denoted by the black boxes: Region 1 (20° N-50° N and 120° E-140° E), Region 2 (20° N-50° N and 140° E-140° W), and Region 3 (20° N-50° N and 140° W-120° W).

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

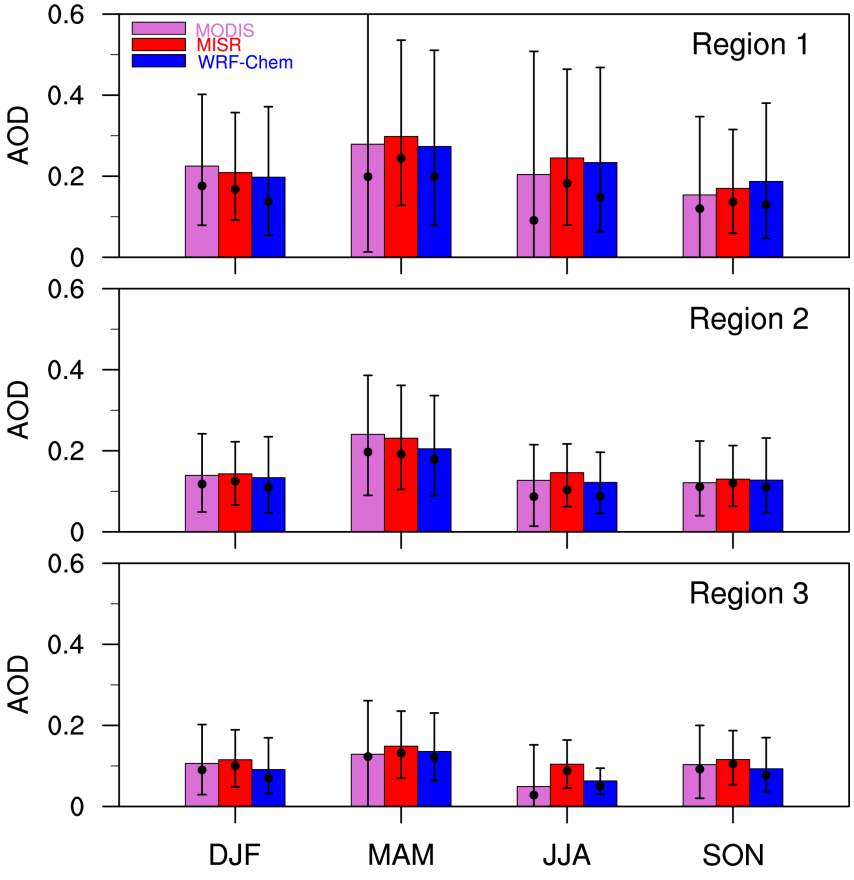

Figure 5 Seasonal mean 550nm AOD from MISR and MODIS retrievals, and the corresponding WRF-Chem simulations averaged for 2010-2014, over the three sub-regions shown in Fig. 4. The values of bars represent the mean. The vertical lines represent 10th and 90th percentile values, and the black dots represent the median values.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

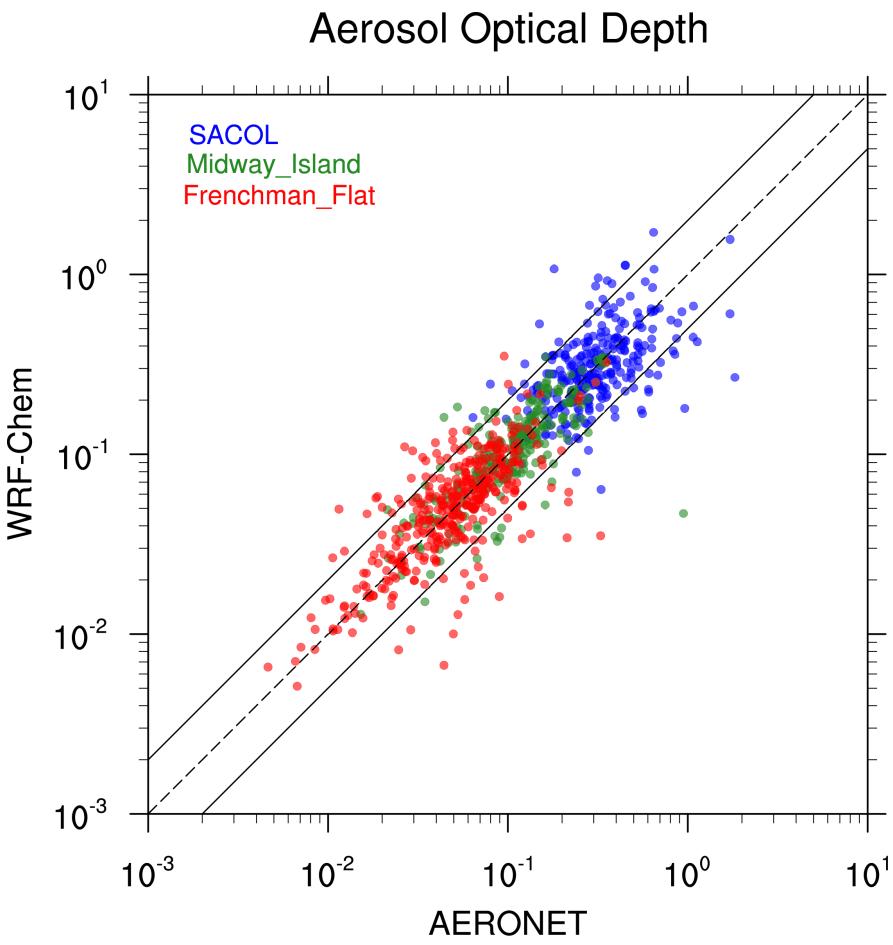

Figure 6 The AERONET observations of daily AOD at 550 nm at the three sites (SACOL, Midway Island, and Frenchman Flat) versus the corresponding WRF-Chem simulations for 2010-2014.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

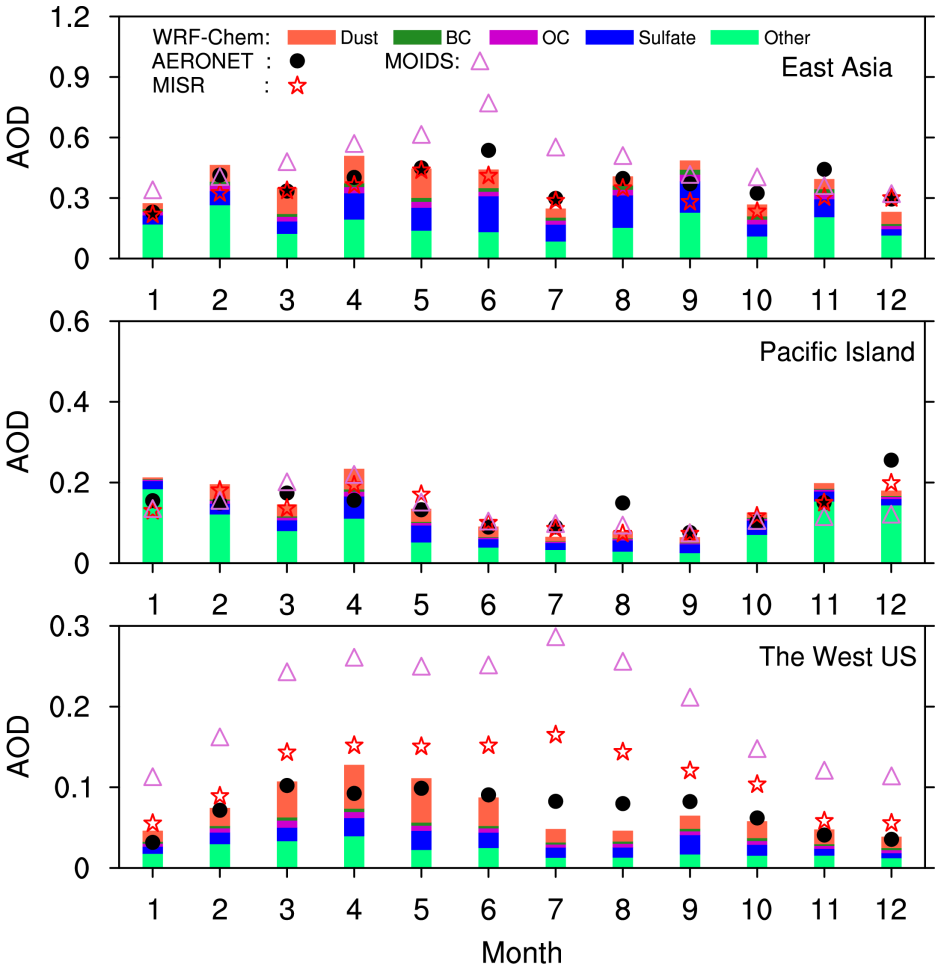

Figure 7 Monthly mean 550nm AOD from AERONET (black dots), MODIS (purple triangles), MISR (red five-pointed stars) and the corresponding WRF-Chem simulations (histogram) averaged for 2010-2014 at the East Asian sites, the Pacific island, and the West U.S. sites as shown in Fig. 1.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

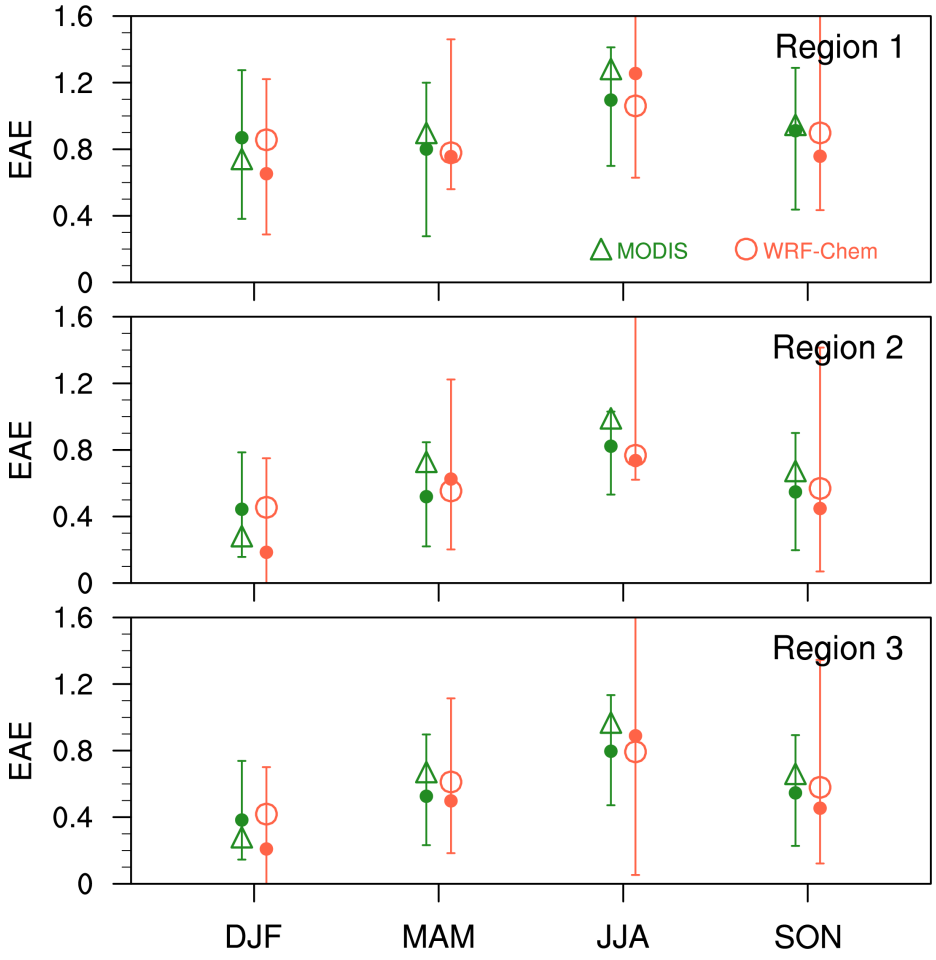

Figure 8 Seasonal mean EAE from the MODIS retrievals and the corresponding WRF-Chem simulations averaged for 2010-2014, over the three sub-regions shown in Fig. 4. The vertical bars represent 10th and 90th percentile values, and the black dots represent the median values.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

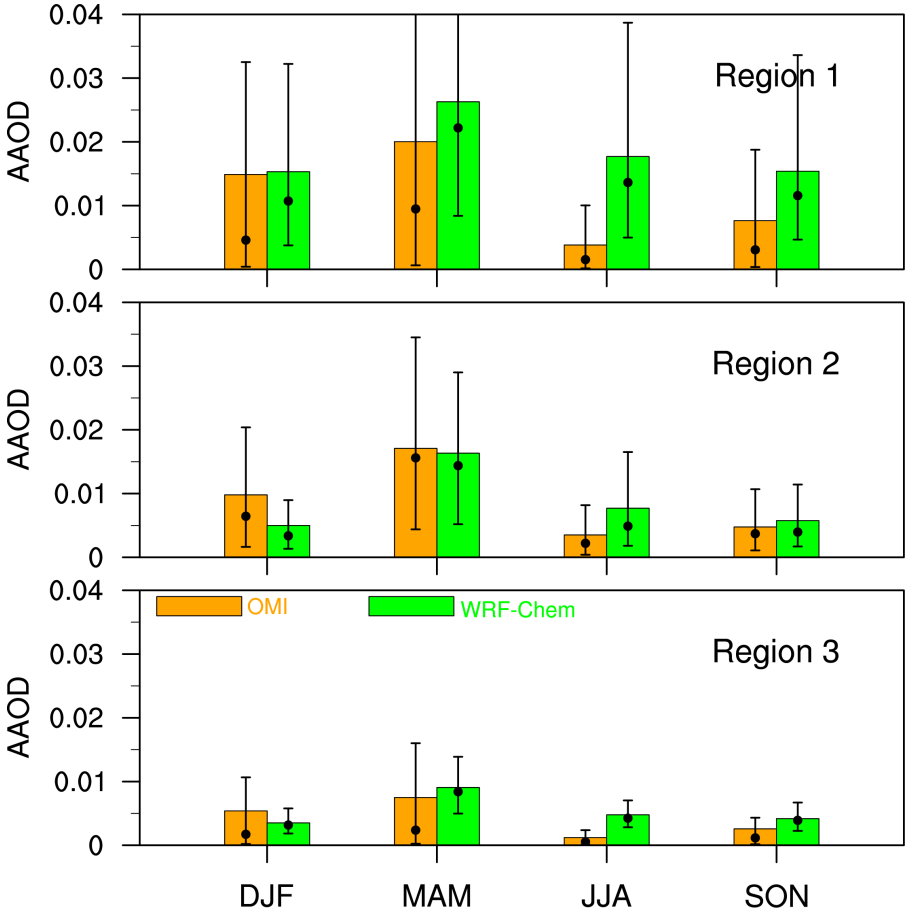

Figure 9 Seasonal mean AAOD at 500 nm from the OMI retrievals and the corresponding WRF-Chem simulations averaged for 2010-2014, over the three sub-regions shown in Fig. 4. The values of bars represent the mean. The vertical lines represent 10th and 90th percentile values, and the black dots represent the median values.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

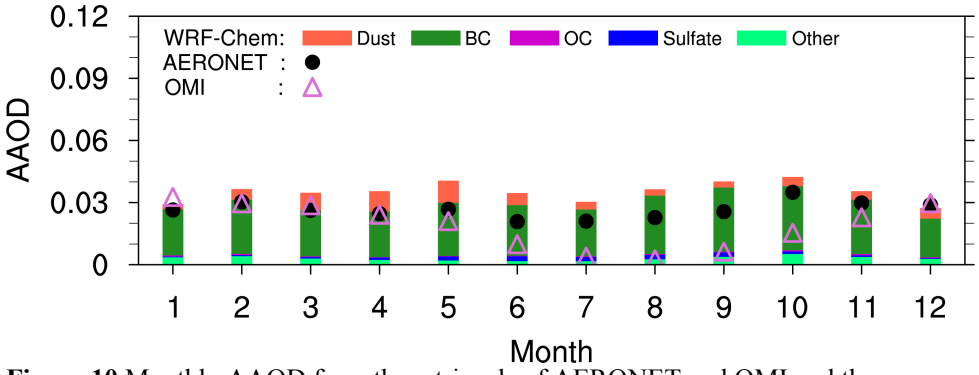

Figure 10 Monthly AAOD from the retrievals of AERONET and OMI and the corresponding WRF-Chem simulations averaged for 2010-2014 over the East Asia sites as shown in Fig. 1.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

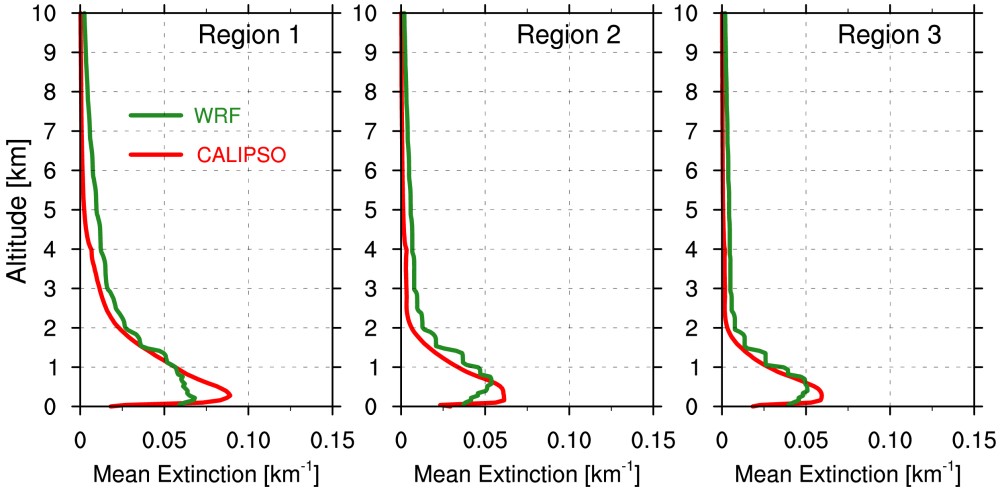

Figure 11 Annual vertical distributions of extinction from CALIPSO observations and the corresponding WRF-Chem simulations averaged for 2010-2014, over the three sub-regions shown in Fig. 4.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

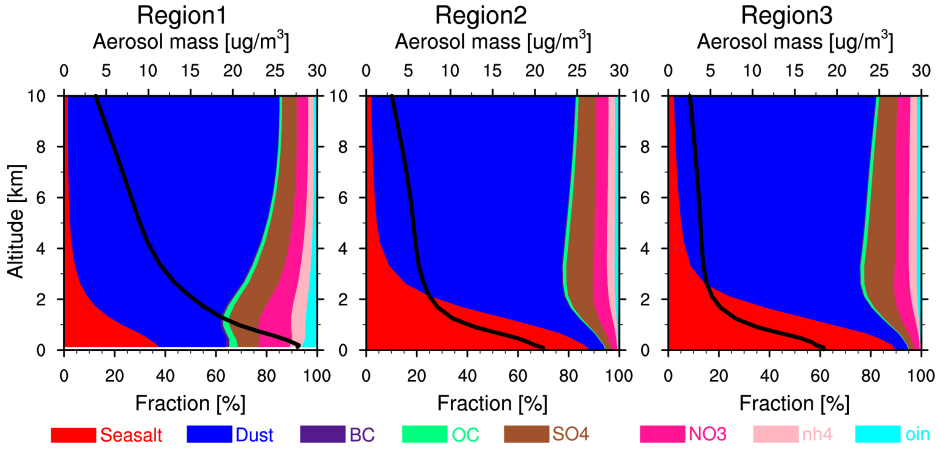

Figure 12 Vertical distributions of mean aerosol mass (black solid line) and its composition fractions (colored shade-contour) from the WRF-Chem simulations averaged for 2010-2014 over three sub-regions as shown in Fig. 4.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

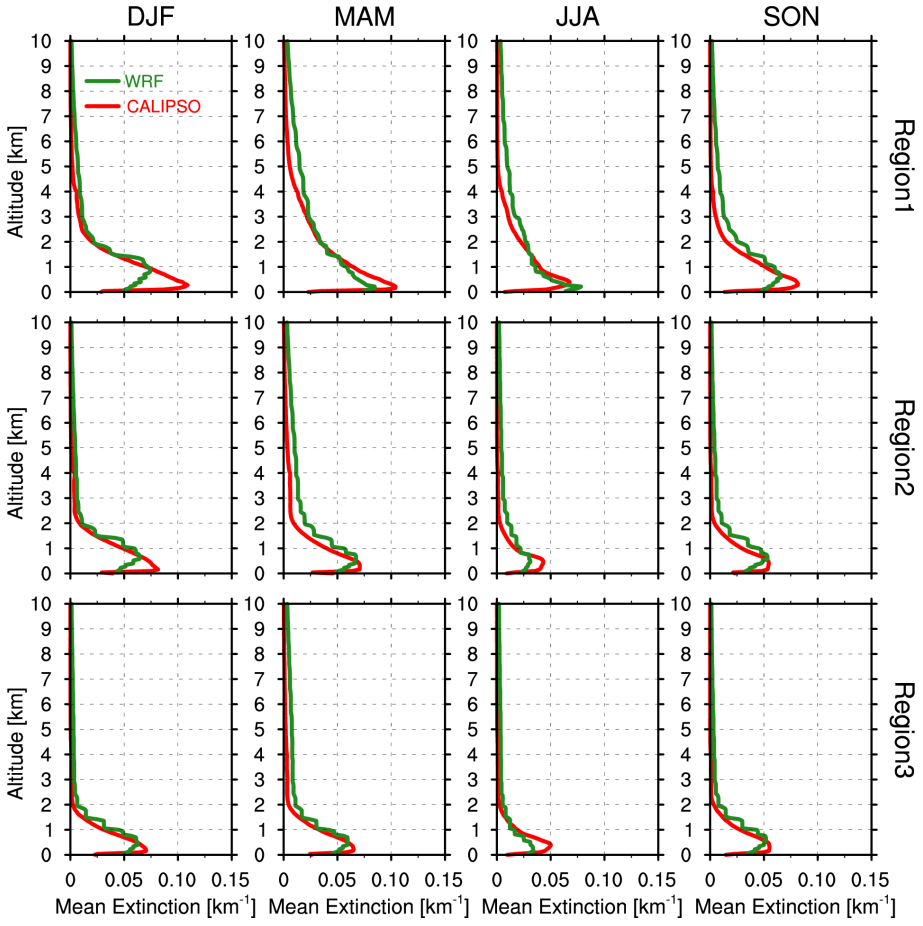

Figure 13 Vertical distributions of seasonal mean aerosol extinction from the CALIPSO retrievals and the corresponding WRF-Chem simulations averaged for 2010-2014 over three sub-regions as shown in Fig. 4.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions
Open Access

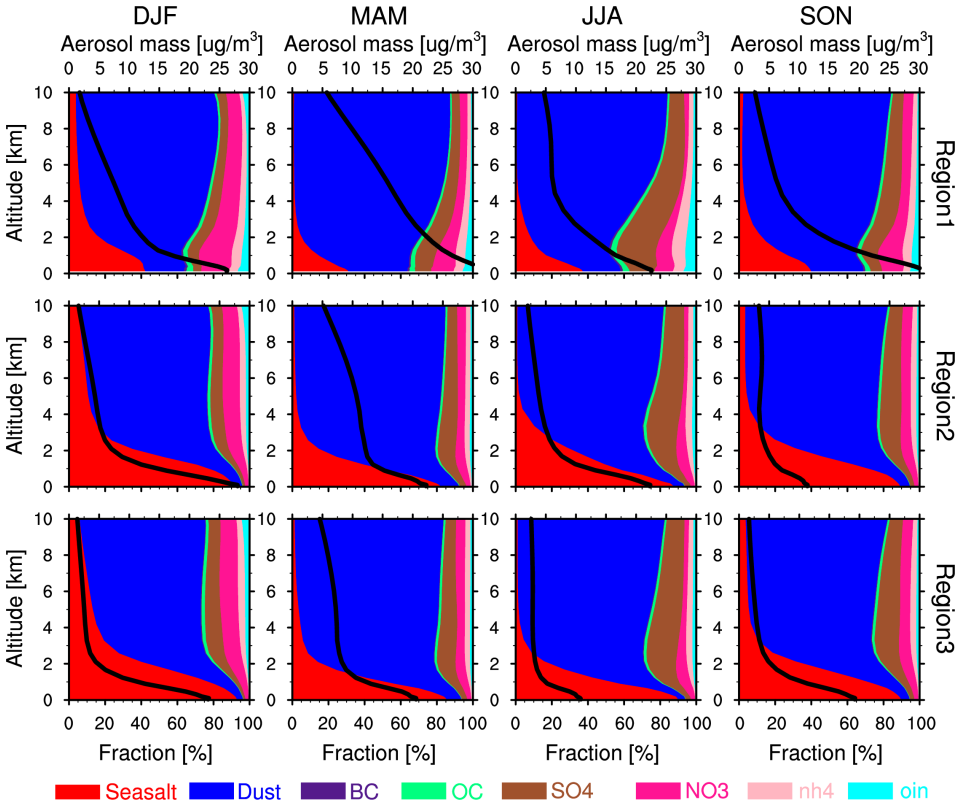

Figure 14 Vertical distributions of seasonal mean aerosol mass (black solid line) and its composition fraction (colored shade-contour) from the WRF-Chem simulations averaged for 2010-2014 over three sub-regions as shown in Fig. 4.

Geosci. Model Dev. Discuss., doi:10.5194/gmd-2015-248, 2016
Manuscript under review for journal Geosci. Model Dev.
Published: 19 January 2016
© Author(s) 2016. CC-BY 3.0 License.

Geoscientific Model Development Discussions

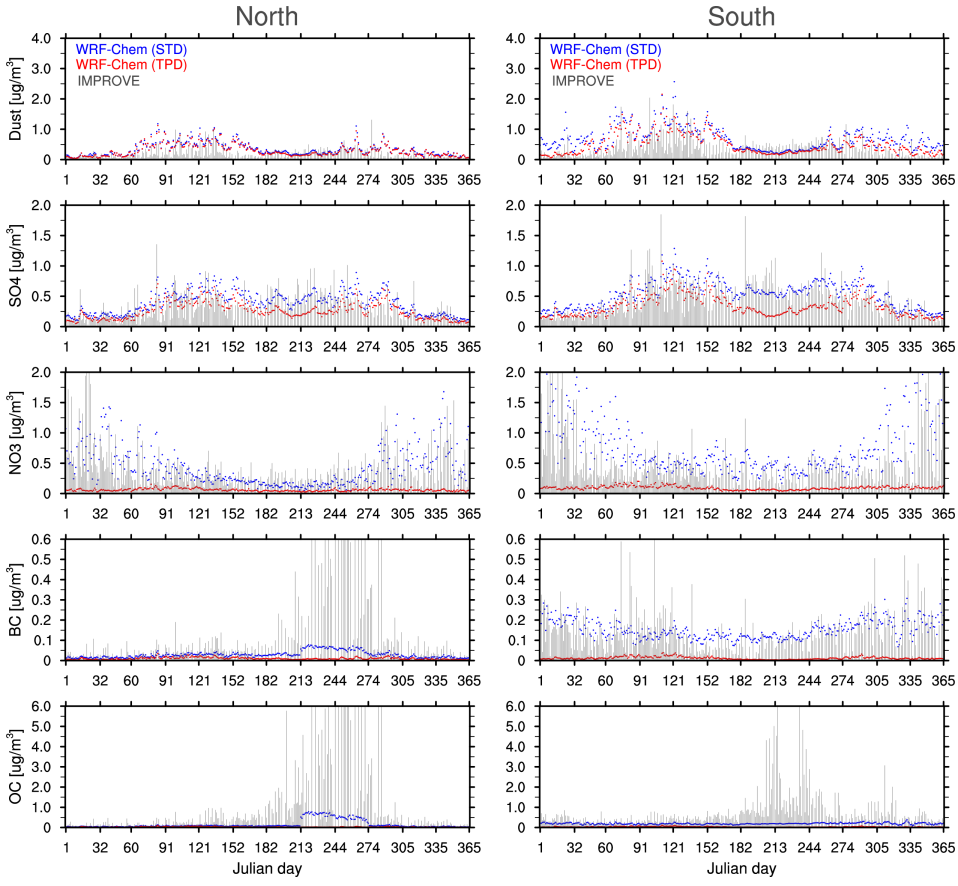

Figure 15 Daily mass concentrations of fine-mode ($PM_{2.5}$) dust, sulfate, nitrate, BC, and OC averaged for 2010-2014 at the IMPROVE sites over the Northwest and Southwest US (shown in Fig. 1) from the IMPROVE observations (vertical gray bars) and the corresponding WRF-Chem standard simulations (STD; blue dots) and the sensitivity simulations without North American emissions (TPD; red dots).