WRF-Chem simulation of aerosol seasonal variability in the San Joaquin Valley

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4	Longtao Wu ¹ , Hui Su ¹ , Olga V. Kalashnikova ¹ , Jonathan H. Jiang ¹ , Chun Zhao ² ,
5	Michael J. Garay ¹ , James R. Campbell ³ and Nanpeng Yu ⁴

6 1. Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA

7 2. School of Earth and Space Sciences, University of Science and Technology of China,

Hefei, Anhui, C	hina
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- 3. Naval Research Laboratory, Monterey, CA, USA
- 10 *4. University of California, Riverside, Riverside, CA, USA*

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17 E-mail: Longtao.Wu@jpl.nasa.gov

¹⁶ Corresponding author address: Longtao Wu, 4800 Oak Grove Dr., Pasadena, CA 91109

18 Highlights:

The WRF-Chem simulation successfully captures aerosol variations in the cold season in the
 San Joaquin Valley (SJV), but has poor performance in the warm season.

- 21 2. High resolution model simulation can better resolve inhomogeneous distribution of
 22 anthropogenic emissions in urban areas, resulting in better simulation of aerosols in the cold
 23 season in the SJV.
- Observations show that dust is a major component of aerosols in the SJV, especially in the
 warm season. Poor performance of the WRF-Chem model in the warm season is mainly due
 to misrepresentation of dust emission and vertical mixing.

27 Abstract

28 WRF-Chem simulations of aerosol seasonal variability in the San Joaquin Valley (SJV), 29 California are evaluated by satellite and in-situ observations. Results show that the WRF-Chem 30 model successfully captures the distribution, magnitude and variation of SJV aerosols during the 31 cold season. However, aerosols are not well represented in the warm season. Aerosol simulations 32 in urban areas during the cold season are sensitive to model horizontal resolution, with better 33 simulations at 4 km resolution than at 20 km resolution, mainly due to inhomogeneous distribution 34 of anthropogenic emissions and better represented precipitation in the 4 km simulation. In rural 35 areas, the model sensitivity to grid size is rather small. Our observational analysis reveals that dust is a primary contributor to aerosols in the SJV, especially during the warm season. Aerosol 36 37 simulations in the warm season are sensitive to parameterization of dust emission in WRF-Chem. 38 The GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport) dust scheme 39 produces very little dust in the SJV while the DUSTRAN (DUST TRANsport model) scheme 40 overestimates dust emission. Vertical mixing of aerosols is not adequately represented in the model 41 based on CALIPSO (Cloud-Aerosol Lidar and Infrared pathfinder Satellite Observation) aerosol 42 extinction profiles. Improved representation of dust emission and vertical mixing in the boundary layer are needed for better simulations of aerosols during the warm season in the SJV. 43

45 **1. Introduction**

46 The San Joaquin Valley (SJV) in the southern portion of the California Central Valley is surrounded by coastal mountain range to the west and the Sierra Nevada range to the east. With 47 48 cool wet winters and hot dry summers, the unique natural environment makes SJV one of the most 49 productive agricultural regions in the world (SJV APCD, 2012 and references therein). However, 50 SJV is also one of the most polluted regions in US due to its unique geographical location. Frequent 51 stagnant weather systems are conducive to air pollution formation, while the surrounding 52 mountains block air flow and trap pollutions. Large seasonal and spatial variation of aerosol 53 occurrence and distribution are observed in the SJV. Although significant progress made to 54 improving local air quality in past decades has been achieved through strong emission controls, 55 PM2.5 (particulate matter with diameter $\leq 2.5 \,\mu$ m) concentrations in the SJV remain well above 56 the national ambient air quality standards (NAAQS) threshold of 12 µg m⁻³ on an annual basis and $35 \mu g m^3$ on daily basis, occurring mainly during the cold season. Improved understanding of the 57 58 aerosol variability and impacts is needed to provide further guidance for emission control strategies 59 in the SJV.

Air quality models are a useful tool to understanding the formation and evolution of aerosols and their impacts on air quality, weather and climate. However, it is quite challenging to accurately simulate aerosol properties (Fast et al., 2014). Fast et al. (2014) summarized the factors contributing to the errors in regional-scale modeling of aerosol properties. They include 1) emission sources; 2) meteorological parameterizations; 3) representation of aerosol chemistry; 4) limited understanding of the formation processes of secondary organic aerosol (SOA); 5) spatial resolution; and 6) boundary conditions. 67 As one of the advanced regional air quality models available presently to the community, the Weather Research and Forecasting model with Chemistry (WRF-Chem) has been widely used 68 69 to study aerosols and their impacts on regional air quality, weather and climate (e.g., Misenis and 70 Zhang, 2010; Zhang et al., 2010; Zhao et al., 2010; 2013a, 2013b; 2014; Gao et al., 2011; Wu et 71 al., 2011a, 2011b, 2013; Fast et al., 2012, 2014; Scarino et al., 2014; Tessum et al., 2015; Campbell 72 et al., 2016; Hu et al., 2016). For example, Fast et al. (2014) showed that WRF-Chem simulations 73 at 4 km horizontal resolution captured the observed meteorology and boundary layer structure over 74 California in May and June of 2010 and the spatial and temporal variations of aerosols were 75 reasonably simulated. Aerosol simulations by WRF-Chem are usually sensitive to both local 76 emission and long-range transport of aerosols from the boundary conditions provided by the global 77 Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). With a similar model 78 set-up, Zhao et al. (2013b) conducted a one-year simulation at 12 km horizontal resolution and 79 found that the WRF-Chem model represented the observed seasonal and spatial variation of 80 surface particulate matter (PM) concentration over California. However, underestimation of 81 elemental carbon (EC) and organic matter (OM) were noticed in the model simulation, with weak 82 sensitivity to horizontal resolution.

In this study, we focus on simulating aerosol seasonal variability in the SJV, California using similar model configurations as that used in Zhao et al. (2013b) and Fast et al. (2014). This paper serves as the first step for future investigation of the aerosol impact on regional climate and the water cycle in California. Previous studies have demonstrated that aerosols are better simulated at higher model resolution (Misenis and Zhang et al., 2010; Qian et al., 2010; Stround et al., 2011; Fountoukis et al., 2013). However, most regional climate studies are still performed with coarse model resolutions (on the order of 10 km) due to the availability of computational resources. This

study will investigate the sensitivity of aerosol simulations to horizontal resolution and identify optimal model physical choices for reasonable representation of aerosol variabilities in the SJV.

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92 Another application of air quality modeling is to provide initial *a priori* fields for remote 93 sensing retrievals. The WRF-Chem model has been proposed as an input for retrieval algorithms 94 to be developed for the recently-selected NASA (National Aeronautics and Space Administration) 95 MAIA (Multi-Angle Imager for Aerosols) mission, which aims to map PM component 96 concentrations in major urban areas (including the SJV, a testbed for the MAIA retrieval algorithm 97 development). A significant challenge for aerosol remote sensing in retrieving spatial information 98 on specific aerosol types, especially near the surface, is caused by the lack of information on the 99 vertical distribution of aerosols in the atmospheric column and limited instrument sensitivity to 100 aerosol types over land. The WRF-Chem model will be used to provide near-real-time estimation 101 of particle properties, aerosol layer heights, and aerosol optical depths (AOD) to constrain the 102 instrument-based PM retrievals. A reasonable estimate of aerosol properties from WRF-Chem is 103 critical to ensuring retrieval speed and quality. Considering the sensitivity of WRF-Chem 104 simulations to various factors such as initial and boundary conditions, model parameterizations 105 and emission sources (e.g., Wu and Petty, 2010; Zhao et al., 2010, 2013a, 2013b; Wu et al., 2011a, 2015; Fast et al., 2014; Campbell et al., 2016; Morabito et al., 2016), careful model evaluations 106 107 are needed before the simulations can be used operationally for remote sensing retrievals. Thus, 108 this study is important for the development of MAIA retrieval algorithms, critical to the success 109 of the MAIA mission.

110 This paper is organized as follows. Section 2 describes observational datasets used for 111 model evaluation. Section 3 provides the description of the WRF-Chem model and experiment setup. Model simulations and their comparison with observations are discussed in section 4.Section 5 presents the conclusions.

114 **2. Observations**

115 2.1 Column-integrated Aerosol Optical Properties

116 AOD is a measure of column-integrated light extinction by aerosols and a proxy for total 117 aerosol loading in the atmospheric column. The Aerosol Robotic Network (AERONET) provides 118 ground measurements of AOD every 15 minutes during daytime under clear skies (Holben et al., 119 1998), with an accuracy approaching ± 0.01 (Eck et al., 1999; Holben et al., 2001; Chew et al., 120 2011). The monthly level 2.0 AOD product with cloud screening and quality control is used in this 121 study. Ångström exponent (AE) is an indicator of aerosol particle size. Small (large) AE values are generally associated with large (small) aerosol particles (Ångström, 1929; Schuster et al., 122 123 2006). The AE between 0.4 µm and 0.6 µm is derived from AERONET observed AODs, and is 124 used to evaluate the model-simulated AE. For comparison with simulated AOD, AERONET AOD 125 is interpolated to 0.55 µm from 0.50 µm and 0.675 µm using the AE. In the SJV, only one 126 AERONET station at Fresno, CA (36.79°N, 119.77°W) has regular observations throughout the 127 California water year 2013 (WY2013) from October 2012 to September 2013.

The Multiangle Imaging Spectroradiometer (MISR) (Diner et al., 1998) instrument onboard the Terra satellite has provided global coverage of AOD once a week since December 130 1999. The standard MISR retrieval algorithm provides AOD observations at 17.6 km resolution 131 using 16x16 pixels of $1.1 \text{ km} \times 1.1 \text{ km}$ each. About 70% of MISR AOD retrievals are within 20% 132 of the paired AERONET AOD, and about 50% of MISR AOD falls within 10% of the AERONET 133 AOD, except in dusty and hybrid (smoke+dust) sites (Kahn et al., 2010). We use version 22 of 134 Level 3 monthly AOD product at 0.5° resolution in this study.

135 2.2 Surface Mass Concentration

136 Surface PM_{2.5} speciation and PM₁₀ (particulate matter with diameter $\leq 10 \mu m$) data are 137 routinely collected by two national chemical speciation monitoring networks: Interagency 138 Monitoring of Protected Visual Environments (IMPROVE) and the $PM_{2,5}$ National Chemical 139 Speciation Network (CSN) operated by Environmental Protection Agency (EPA) (Hand et al. 140 2011; Solomon et al., 2014). IMPROVE collects 24-h aerosol speciation every third day at mostly 141 rural sites since 1988. The same frequency of aerosol speciation dataset was collected at EPA CSN 142 sites in urban and suburban areas since 2000. The observed organic carbon is converted to OM by 143 multiplying by 1.4 (Zhao et al., 2013b; Hu et al., 2016). Some precursors of aerosol pollutions 144 **EPA** (such as NO_2 and SO_2) are observed hourly by (data available at: https://aqsdr1.epa.gov/aqsweb/aqstmp/airdata/download_files.html) and are used in this study. 145 146 Selected IMPROVE and EPA CSN sites used in this study are shown in Figure 1a.

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2.3 Aerosol Extinction Profile

148 The aerosol extinction coefficient profile reflects the attenuation of the light passing 149 through the atmosphere due to the scattering and absorption by aerosol particles as a function of 150 range. Version 3 Level 2 532 nm aerosol extinction profiles derived from Cloud-Aerosol Lidar 151 with Orthogonal Polarization (CALIOP) backscatter profiles collected onboard the Cloud-Aerosol 152 Lidar and Infrared pathfinder Satellite Observation (CALIPSO) satellite are used (Omar et al., 153 2009; Young and Vaughan, 2009). Seasonal mean profiles are derived for WY2013 based on the 154 methodology outlined in Campbell et al. (2012), whereby quality-assurance protocols are applied 155 to individual profiles before aggregating and averaging the data. We highlight that no individual profiles are included in the averages if the CALIOP Level 2 retrieval failed to resolve any 156 157 extinction within the column, a potential issue to create bias that has recently been described by

Toth et al. (2017). Level 2 532 nm aerosol extinction data classify aerosols into 6 types: clean marine, dust, polluted continental, clean continental, polluted dust and smoke. Dust and polluted dust are distinguished in the averages in this study for their contribution to total extinction and the vertical profile seasonally in the SJV.

162 **2.4 Meteorology**

163 AIRS (Atmospheric Infrared Sounder) onboard the Aqua satellite (Susskind et al., 2003; 164 Divakarla et al., 2006) has provided global coverage of the tropospheric temperature and moisture 165 at approximately 01:30 and 13:30 local time since 2002. AIRS retrievals have root-mean-squared 166 (RMS) error of ~1 K for temperature and ~15% for water vapor (Divakarla et al., 2006). Level 3 167 monthly temperature and moisture retrievals (version 6) at $1^{\circ} \times 1^{\circ}$ grid are used in this study. Vertical gradient of equivalent potential temperature (θ_e) marks atmospheric stability and is 168 169 computed from temperature and moisture profiles observed by AIRS. Vertical profiles from the 170 European Center for Medium-Range Weather Forecasts Interim Re-Analysis (ERA-Interim; Dee 171 et al., 2011) are also used for comparison. Surface observations, including air temperature, relative 172 humidity (RH) and wind speed, are routinely collected at the California Irrigation Management 173 Information System (CIMIS; http://www.cimis.water.ca.gov/). Precipitation used in this study is 174 the Climate Prediction Center (CPC) Unified Gauge-Based Analysis of Daily Precipitation product at $0.25^{\circ} \ge 0.25^{\circ}$ resolution. 175

176 **3. Model Description and Experiment Setup**

The WRF-Chem model Version 3.5.1 (Grell et al., 2005) updated by Pacific Northwest National Laboratory (PNNL) is used in this study (Zhao et al., 2014). This study uses the CBM-Z (carbon bond mechanism) photochemical mechanism (Zaveri and Peters, 1999) coupled with the sectional-bin MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol 181 scheme (Zaveri et al., 2008) as the chemical driver. The major components of aerosols (nitrate, 182 ammonium, EC, primary OM, sulfate, sea salt, dust, water and other inorganic matter) as well as 183 their physical and chemical processes are simulated in the model. For computational efficiency, 184 aerosol particles in this study are partitioned into four-sectional bins with dry diameter within 185 0.039-0.156 µm, 0.156-0.625 µm, 0.625-2.5 µm, and 2.5-10.0 µm. Zhao et al. (2013a) compared 186 the impact of aerosol size partition on dust simulations. It showed that the 4-bin approach 187 reasonably produces dust mass loading and AOD compared with the 8-bin approach. The size 188 distribution of the 4-bin approach follows that of the 8-bin approach with coarser resolution, 189 resulting in $\pm 5\%$ difference on the ratio of PM_{2.5}-dust/PM₁₀-dust in dusty regions (more large 190 particles and less small particles). Dust number loading and absorptivity are biased high in the 4bin approach compared with the 8-bin approach. 191

192 Aerosols are considered to be spherical and internally mixed in each bin (Barnard et al., 193 2006; Zhao et al., 2013b). The bulk refractive index for each particle is calculated by volume 194 averaging in each bin. Mie calculations as described by Ghan et al. (2001) are used to derive 195 aerosol optical properties (such as extinction, single-scattering albedo, and the asymmetry 196 parameter for scattering) as a function of wavelength. Aerosol radiation interaction is included in 197 the shortwave and longwave radiation schemes (Fast et al., 2006; Zhao et al., 2011). By linking 198 simulated cloud droplet number with shortwave radiation and microphysics schemes, aerosol 199 cloud interaction is effectively simulated in WRF-Chem (Chapman et al., 2009). Aerosol snow 200 interaction is implemented in this version of WRF-Chem (Zhao et al., 2014) by considering aerosol 201 deposition on snow and the subsequent radiative impacts through the SNICAR (SNow, ICe, and 202 Aerosol Radiative) model (Flanner and Zender, 2005, 2006).

203 The model simulations start on 1 September 2012 and run continuously for 13 months. 204 With the first month used for the model spin-up, our analysis focuses on WY2013 from October 205 2012 to September 2013. The model is configured with 40 vertical levels and a model top at 50 206 hPa. The vertical resolution from the surface to 1 km gradually increases from 28 m to 250 m. The 207 model center is placed at 38° N, 121° W, with 250×350 grid points at 4 km horizontal resolution 208 (referred to as "4km" hereafter; Table 1), covering California and the surrounding area. To test the 209 sensitivity of the aerosol simulations to horizontal resolution, one simulation with the same model 210 settings and domain coverage is conducted at 20 km horizontal resolution (referred to as "20km" 211 hereafter).

212 The physics parameterizations used in the simulations include the Morrison double-213 moment microphysics scheme (Morrison et al., 2009), Rapid Radiative Transfer Model for General 214 circulation model (RRTMG) shortwave and longwave radiation schemes (Iacono et al., 2008), 215 Community Land Model (CLM) Version 4 land surface scheme (Lawrence et al., 2011). The 216 Yonsei University (YSU) planetary boundary layer (PBL) scheme (Hong et al., 2006) is used in 217 all of the simulations, except one sensitivity experiment that uses the ACM2 (Asymmetric 218 Convective Model with non-local upward mixing and local downward mixing; Pleim, 2007) PBL 219 scheme (referred to as "20km_P7" hereafter, Table 1). Previous studies showed that both YSU and 220 ACM2 schemes have good performance in simulating boundary layer properties (e.g., Hu et al., 221 2010; Xie et al., 2012; Cuchiara et al., 2014; Banks and Baldasano, 2016; Banks et al., 2016; Chen et al., 2017). Subgrid convection, convective transport of chemical constituents and aerosols, and 222 223 wet deposition from subgrid convection are parameterized using the Grell 3D ensemble cumulus 224 scheme (Grell and Devenyi, 2002) in the 20 km simulations while convective processes are 225 resolved in the 4 km simulations. The ERA-Interim reanalysis serves as initial and boundary

meteorological conditions for WRF-Chem. The MOZART-4 global chemical transport model (Emmons et al., 2010) is used for initial and boundary chemical conditions. Fast et al. (2014) found that the MOZART-4 model overestimates aerosols in the free troposphere over California, which is also found in one of our sensitivity experiments ("20km_BC1" in the supplementary). Following Fast et al. (2014), the chemical initial and boundary conditions from MOZART-4 are divided by two in all simulations except 20km_BC1.

232 Anthropogenic emissions are provided by US EPA 2005 National Emissions Inventory 233 (NEI05), with area-type emissions on a structured 4-km grid and point-type emissions at specific 234 latitude and longitude locations (US EPA, 2010). Nineteen gases (including SO₂, NO, NH₃ etc.) 235 are emitted, and aerosol emissions include SO₄, NO₃, EC, organic aerosols, and total PM_{2.5} and 236 PM₁₀ masses. Anthropogenic emissions are updated every hour to account for diurnal variability, 237 while its seasonal variation is not considered in the simulations. A sensitivity experiment with 2011 NEI emissions ("20km_NEI11" in the supplementary) does not produce significantly 238 239 different results from the 2005 NEI emissions. Biogenic emissions are calculated online using the 240 Model of Emissions of Gases and Aerosols from Nature (MEGAN) model (Guenther et al., 2006). 241 Biomass burning emissions are obtained from the Global Fire Emissions Database version 2.1, with eight-day temporal resolution (Randerson et al., 2007) and updated monthly. Sea salt 242 243 emissions are derived from the PNNL-updated sea salt emission scheme that includes the 244 correction of particles with radius less than 0.2 µm (Gong et al., 2003) and dependence on sea 245 surface temperature (Jaeglé et al., 2011).

Following Zhao et al. (2013b), dust emission is computed from the GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport) dust scheme (Ginoux et al., 2001) in the 20km and 4km simulations. The GOCART dust scheme estimates the dust emission flux *F* as

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$$F = CSs_p u_{10m}^2 (u_{10m} - u_t)$$

where *C* is an empirical proportionality constant, *S* is a source function for potential wind erosion that is derived from $1^{\circ} \times 1^{\circ}$ GOCART database (Freitas et al., 2011), s_p is a fraction of each size class dust in emission, u_{10m} is 10-m wind speed and u_t is a threshold speed for dust emission.

As shown later, a significant amount of dust is observed in the SJV, whereas the GOCART dust scheme produces little dust. Two sensitivity experiments at 20 km and 4 km horizontal resolution (hereafter referred to as "20km_D2" and "4km_D2", respectively) are conducted by switching the dust emission scheme to the DUST TRANsport model (DUSTRAN) scheme (Shaw et al., 2008). The DUSTRAN scheme estimates *F* as

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$$F = \alpha C u_*^4 (1 - \frac{f_w u_{*t}}{u_*})$$

where *C* is an empirical proportionality constant, α is the vegetation mask, u_* is the friction velocity, u_{*t} is a threshold friction velocity and f_w is the soil wetness factor. The *C* value in both GOCART and DUSTRAN is highly tunable for different regions. The original *C* values, 1.0 µg s² m⁻⁵ in GOCART (Ginoux et al., 2001) and 1.0×10^{-14} g cm⁻⁶ s⁻³ in DUSTRAN (Shaw et al., 2008), are used in this study.

4. Model Simulation Results

Shown in Fig. 1a, our model domain includes three urban sites (Fresno, Bakersfield and Modesto) and two rural sites (Pinnacles and Kaiser) where surface measurements of aerosols are available. Because aerosols properties and model performance are similar at all urban sites, our discussion is focused on the results at Fresno and the simulations for other urban sites are provided in the supplementary materials. Model simulations in the rural areas are presented in the last subsection.

4.1 Sensitivity to Horizontal Resolution

272 Figure 1 features daily mean anthropogenic PM_{25} emission rates used in the 20km and 273 4km simulations, respectively. Although both emission rates are derived from the 4 km NEI05 274 dataset, localized high emission rates with sharp gradients are evident in urban areas from the 4km 275 simulation (Fig. 1b). The 20km simulation exhibits lower emission rates at the urban areas with 276 weaker gradients due to the reapportionment process (Fig. 1a). As precipitation is an important 277 process that removes aerosols, we examine the simulated precipitation for the 20km and 4km runs 278 and find that the 20km simulation produces 51% more precipitation, although the domain averaged 279 precipitation is lower in the 20km run than the 4km run (Fig. 2a).

280 Consistent with higher emission rates and lower precipitation at Fresno, the 4km run 281 simulates higher AOD than the 20km run in the cold season (October-November-December and 282 January-February-March; OND and JFM in Fig. 3). Averaged over a broad area encompassing 283 Fresno and Bakersfield, the most polluted region in the SJV (red box in Fig. 1a), the AOD is 0.090 284 in the 4km and 0.073 in the 20km, a 23% difference. Compared to the MISR observations, the 285 4km simulation reproduces the spatial distribution and magnitude of AOD in the cold season. 286 However, the AOD difference between the 20km and 4km runs is small in the warm season (April-287 May-June and July-August-September; AMJ and JAS in Fig. 3), and both runs underestimate AOD 288 by ~50% with respect to the MISR observations.

Comparing the point values at Fresno in the 4km and 20km simulations (Fig. 4a), we find similar results: the 4km AOD is closer to the AERONET measurements and is about 23% higher than that in the 20km run during the cold season, while both runs are biased low in AOD during the warm season. The different model sensitivities to horizontal resolution between the cold and warm seasons suggest that the dominant aerosol sources may be different for the two seasons. We will elaborate upon the aerosol composition in the following section. MISR and AERONET 295 observations display weak seasonal AOD variation in the SJV and at Fresno, respectively, which 296 is not well represented in the 20km and 4km simulations (Fig. 3 and 4a).

297 Aside from AOD, significant seasonal variability of AE (Fig. 4b) is shown at Fresno. AE 298 exhibits a maximum about 1.50 in January and a minimum of 0.98 in April, suggesting relatively 299 small particles in the winter and large particles in the spring. A relatively large AE value of 1.40 300 (corresponding to small particles) is observed in July, possibly related to the wild fires in late July 301 in the SJV. WRF-Chem captures the seasonal variability of the AE well, with a correlation of 0.90 302 in both the 20km and 4km simulations. The magnitude of AE is also approximately simulated in 303 the cold season, with a mean of 1.15 (1.20) in the 20km (4km) runs compared to 1.33 in the 304 observation. However, the simulated AE is underestimated by $\sim 30\%$ in the warm season, 305 indicating that the simulated particle size is biased high during this period.

306 Significant seasonal variability of PM_{2.5} is observed in the SJV urban areas (Fig. 5a and Supplementary Fig. 4a and 5a). PM_{2.5} at Fresno peaks in January (26.18 µg m⁻³) and reaches a 307 308 minimum of 7.03 μ g m⁻³ in June, with an annual nonattainment value of 12.64 μ g m⁻³ (Fig. 5a). 309 Both the 20km and 4km runs approximately capture the observed seasonal variability of $PM_{2.5}$, 310 with a correlation around 0.90 (Table 2). In the cold season, the 4km simulation overestimates PM_{2.5} by 27% while the 20km simulation exhibits a low bias of 19% compared with IMPROVE 311 312 observations at Fresno (Table 3). The 4km simulation of PM_{10} is in good agreement with 313 IMPROVE in the winter (December, January and February), but has significant low biases of 314 between 30% and 85% in other months (Fig. 5b). The 20km simulation underestimates PM_{10} 315 throughout WY2013.

316 $PM_{2.5}$ is a mixture of nitrate (NO₃), ammonia (NH₄), OM, EC, sulfate (SO₄), dust and other 317 aerosols. High concentrations of $PM_{2.5}$ are primarily the result of NO₃ at Fresno (Fig. 5c). Both simulations produce the seasonal variability of NO₃ with a correlation of 0.94, but high bias of 17% (75%) is found in the 20km (4km) simulations during the cold season. As one precursor of NO₃, NO₂ is underestimated by 43% in the 20km run (Fig. 6a). The overestimation in NO₃ and underestimation in NO₂ suggest that the precursor emissions may not the reason for the high biases in NO₃. NH₄ shows a similar performance to NO₃, with an overestimation by 38% (111%) in the 20km (4km) runs during the cold seasons (Fig. 5d). As shown later in section 4.3, both NO₃ and NH₄ simulations are quite sensitive to the PBL scheme applied.

325 OM, the second largest contributing species to cold season $PM_{2.5}$ in the SJV (Table 3), is 326 significantly underestimated by 82% in the 20km simulation (Fig. 5f). The 4km simulation 327 produces higher OM, but it is still lower than the IMPROVE observations by 63%. The 328 underestimation of OM is expected, because SOA processes are not included in our model 329 infrastructure. Fast et al. (2014) used the simplified two-product volatility basis set 330 parameterization to simulate equilibrium SOA partitioning in WRF-Chem although SOA was still 331 underestimated in their simulation. It remains ongoing research how to correctly represent SOA 332 processes in regional climate models.

Both the 20km and 4km simulations reproduce the seasonal variability of EC, with a correlation of 0.98 between the modeled and observed time series (Table 2). The 20km simulation underestimates EC by 52% (16%) in the cold (warm) season (Fig. 5e and Table 3). The 4km simulated EC (1.12 μ g m⁻³) exhibits good agreement with IMPROVE (1.08 μ g m⁻³) in the cold season, but overestimates EC by 53% in the warm season.

The seasonal variability of SO_4 at Fresno is very different from other $PM_{2.5}$ species. It peaks in May at 1.35 µg m⁻³ and reaches the minimum of 0.67 µg m⁻³ in August (Fig. 5g). The 20km simulated SO_4 exhibits good correlation of 0.63 with the observation (Table 2), but is biased low 341 by 28% to 63% throughout WY2013 (Fig. 5g). Although the observed SO_2 , the precursor of SO_4 , 342 has approximately similar seasonal variation to the observed SO_4 (Fig. 6b), the 20km simulated 343 seasonal variability of SO₂ resembles other anthropogenic emissions, with high values in the cold 344 season and low values in the warm season, out of phase with the simulated SO_4 and the observed SO₂. The 4km simulation produces higher SO₄ than the 20km run, resulting in better agreement 345 346 with the observation (0.82 μ g m⁻³ vs. 0.87 μ g m⁻³) during the cold season (Fig. 5g and Table 3). 347 However, the 4km run produces an increase of SO_4 by only 13% comparing to the 20km run in 348 the warm season, resulting in a correlation of -0.16 between the 4km simulation and the 349 observation.

350 To explore the possible cause for the underestimation of SO₄ and SO₂ in the warm season 351 in both the 20km and 4km simulations, we conduct a sensitivity experiment with different chemical 352 boundary conditions from the baseline runs (20km_BC1 in the supplementary). We find that SO_4 353 in the SJV is partly contributed to by marine intrusions (the different chemical boundary conditions 354 between 20km_BC1 and 20km_D2) throughout the year (supplementary Fig. 2g), as pointed out 355 by Fast et al. (2014). Including the marine intrusions, the 20km_BC1 simulated SO₄ tracks the 356 observation at a correlation of 0.78. Doubled chemical boundary conditions in the 20km simulation results in 41% increase in SO₄ at Fresno, with a stronger increase in the warm season. Compared 357 358 to the observed SO₄ of 1.04 μ g m⁻³ in the warm season, the simulated SO₄ of 0.79 μ g m⁻³ in the 359 20km BC1 run is closer to the observation than that simulated in the 20km D2 run (0.53 µg m⁻³). 360 The relative contributions of local emissions and remote transports (as well as other emission 361 sources, such as wild fires) to SO₄ concentrations in different seasons of the SJV require further 362 investigation.

Overall, the 4km simulation produces higher AOD and surface PM than the 20km 363 364 simulation in urban areas of the SJV, especially during the cold season, resulting in better agreement with satellite and surface observations than the 20km simulation. Both the 20km and 365 4km simulations approximately capture the seasonal variability of $PM_{2,5}$ and most of its speciation. 366 However, significant low biases of AOD and PM₁₀ are found during the warm season in both 367 368 simulations. The underestimation also exists in a sensitivity experiment (not shown) with the same 369 model setups except initialized in April, indicating that the identified model biases during the warm 370 season are not caused by potential model drift after a relatively long simulation period. The 371 relatively good performance in simulating $PM_{2.5}$ but not PM_{10} during the warm season suggests 372 that coarse aerosol particle mass (CM; 10 μ m \geq particulate matter with diameter > 2.5 μ m), mainly 373 dust in the SJV, is not properly represented in the model. The impact of dust parameterizations is 374 investigated in the 4km_D2 experiment.

375

4.2 Sensitivity to Dust Scheme

376 Limited amounts of PM_{2.5}_dust (dust with diameter $\leq 2.5 \,\mu$ m) are observed in the SJV cold 377 season, with a minimum of 0.37 μ g m⁻³ in December (Fig. 7a). The amount of PM_{2.5} dust increases 378 in the warm season, with a peak of 3.86 μ g m⁻³ in September. The 4km simulation produces 379 comparable $PM_{2,5}$ dust relative to IMPROVE in the winter, but almost no dust in other months 380 (Fig. 7 and upper panel in Fig. 8). On the other hand, the dust emission rate in the 4km D2 run is 381 significantly higher than the 4km run. We have found that the source function, S, for potential 382 wind erosion in the SJV is set to zero in the $1^{\circ} \times 1^{\circ}$ GOCART dataset used for the 4km simulation 383 (Fig. 9). An updated source function, S, at higher resolution is needed for the GOCART dust 384 scheme to correctly represent dust emissions in the SJV.

385 The 4km_D2 simulation reproduces the amount of PM_{2.5}_dust in OND (Fig. 7a). However, 386 it overestimates $PM_{2.5}$ dust by up to a factor of 3 in the warm season, resulting in an overestimation 387 of PM_{2.5} by 52% (Fig. 7b and Table 3). PM_{2.5} dust is not sensitive to long-range transport (from 388 chemical boundary conditions in the model simulation; Supplementary Fig. 2h). Both the 4km and 389 4km_D2 simulations capture the seasonal variability of PM_{2.5}, but not that of PM₁₀ (Fig. 7c). The 390 magnitude of PM_{10} in the 4km_D2 run is larger than the 4km simulation. PM_{10} in the 4km_D2 run 391 is overestimated in April-May-June (AMJ) but underestimated in July-August-September (JAS), 392 leading to a comparable season mean of $38.12 \ \mu g \ m^{-3}$ with IMPROVE observed $34.82 \ \mu g \ m^{-3}$. The 393 overestimation of AMJ PM₁₀ and PM_{2.5}_dust in the 4km_D2 run is likely associated with the high 394 bias in the simulated wind speed (Fig. 2b).

395 On the relative contribution of different aerosol species, IMPROVE observations at Fresno 396 show that NO₃ is the primary contributor (32.3%) to $PM_{2.5}$ while only 5.3% of $PM_{2.5}$ is dust in the 397 cold season (panel 1 of Fig. 10). Both the 4km and 4km_D2 runs roughly reproduce the relative 398 contributions to PM_{2.5} in the cold season, with an overestimation of NO₃ and NH₄ and an 399 underestimation of OM, consistent with the time series in Fig. 5. Relative contributions of dust to 400 PM_{2.5} are better simulated in the 4km_D2 run (7.3%) than the 4km one (<1.0%). IMPROVE shows 401 that 46.6% of PM_{10} is CM in the cold season (panel 2 of Fig. 10). Both the 4km (6.3%) and 402 4km_D2 (20.6%) runs underestimate the contribution of CM to PM₁₀, mainly in October and 403 November. In the warm season, dust (24.6%) becomes the primary contributor to PM_{2.5} while the 404 contribution from NO₃ decreases to 9.9% in IMPROVE observations (panel 3 of Fig. 10). Almost 405 no PM_{2.5}_dust is simulated in the 4km run while too much PM_{2.5}_dust is produced in the 4km_D2 406 (50.5%) run during the warm season. The relative contribution of CM to PM₁₀ is too small (27.6%)

in the 4km run, while the 4km_D2 run reflects an better relative contribution of 66.3% as compared
to an IMPROVE observed 75.8% (panel 4 of Fig. 10).

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409 AOD simulations are improved in the 4km D2 experiment (Fig. 11), with better agreement 410 found from MISR (Fig. 3) in AMJ. AOD (0.114) in the 4km D2 run is comparable to observations 411 (0.131) in AMJ, but still underestimated by 53% in JAS. Consistent with AOD, the vertical 412 distribution of aerosol extinction is reasonably simulated during the cold season in the WRF-Chem 413 simulations, while large discrepancies are found in the warm season (Fig. 12). As observed by 414 CALIOP at 532 nm, aerosols are confined below 1 km in the cold season and decrease sharply 415 with height. During AMJ, aerosols are well mixed between the surface and the altitude of 1.5 km 416 and then decrease with height gradually. During JAS, the well-mixed aerosol layer is shallower 417 than that in AMJ and the vertical profile of aerosol extinction is in-between the cold season and 418 AMJ. Model simulations roughly capture the "bottom-heavy" structure of the extinction profiles 419 observed by CALIOP especially in the cold season, but significant biases exist. One common 420 problem for all four seasons is the low bias in the boundary layer and high bias in the free 421 atmosphere. Similar discrepancy between the model simulations and CALIOP is shown in other 422 studies (Wu et al., 2011a; Hu et al., 2016). The model does not capture the well-mixed aerosol layer during AMJ. The difference in the aerosol extinction profiles between the 4km and 4km D2 423 424 runs is small during the cold season.

Dust in the boundary layer is a primary factor contributing to aerosol extinction in the SJV, as illustrated by the differences between the bulk seasonal CALIOP mean profile and those excluding the contributions of the dust and polluted dust (CALIOP_nodust) profiles (Fig. 12). Simulated aerosol extinction falls between the two in all seasons, suggesting that dust is the primary factor contributing to the model biases in aerosol extinction. Although a small portion of 430 PM_{25} is dust in the cold season, it contributes to about 50% of total aerosol extinction (Fig. 12a) 431 and 12b). A predominant portion of aerosol extinction in the lower troposphere is contributed by 432 dust in the warm season (Fig. 12c and 12d). There, the 4km D2 simulation produces higher aerosol 433 extinction between 0.3 km and 3 km than the 4km simulation, although it is still lower than 434 CALIOP. The simulated aerosol extinction in the free troposphere is close to or larger than 435 CALIOP, suggesting that aerosols transported from remote areas through chemical boundary 436 conditions (e.g., the differences between the 20km BC1 and 20km D2 runs in Supplementary Fig. 437 3) may not be the major factor contributing to the underestimation of dust between 0.3 km and 3 438 km in the SJV.

Overall, the poor simulations of dust play a dominant role in the low bias of aerosols in the boundary layer during the warm season. Both the GOCART and DUSTRAN dust emission schemes used in this study have difficulties in reproducing dust emissions in the SJV, with an underestimation in GOCART and an overestimation in DUSTRAN (Fig. 7). Improvement on the dust emission schemes is needed for capturing the seasonal variability of aerosols in the SJV.

444

4.3 The Role of Meteorology

The WRF-Chem simulations approximately reproduce the seasonal variations of 445 446 meteorological variables near the surface (correlations > 0.80), including temperature, RH, wind 447 speed and precipitation (Supplementary Fig. 6 and Supplementary Table 1). All of the model simulations exhibit warm and dry biases near surface and in the boundary layer, with cold and wet 448 449 biases in the free atmosphere (Supplementary Fig. 6-8 and Supplementary Table 2). The dry bias 450 in the 4km D2 run is about 10% near the surface throughout WY2013. Due to the relative dry 451 environment (RH<50%) in the warm season, the underestimation of boundary layer aerosol 452 extinction and column-integrated AOD is unlikely caused by the hygroscopic effects (Feingold

453 and Morley, 2003). In the cold season, the surface wind speed is underestimated by 0.67 m s⁻¹ 454 (1.00 m s⁻¹) in the 4km_D2 (20km_D2) runs. In the warm season, the 4km_D2 run overestimates 455 wind speed by 0.78 m s⁻¹, while the 20km D2 run has an underestimation of 0.16 m s⁻¹. These 456 results suggest that wind speed is not a major factor contributing to the low biases of aerosols in 457 the boundary layer between 0.3 km and 3 km. Furthermore, the seasonal variability of precipitation 458 is well captured in the simulations, while the magnitude of precipitation is weaker than the 459 observations during the warm season (Supplementary Table 2). Thus, we conclude that wet 460 removal processes would not be a primary reason for the aerosol biases in the warm season.

461 In the warm season, more aerosols are observed above 1.5 km than in the cold season (Fig. 462 12). A well-mixed layer of aerosols is observed below 1.5 km in AMJ (Fig. 12c), consistent with 463 the unstable lower troposphere below 1.5 km shown in AIRS and ERA-Interim (Fig. 13c). The 464 WRF-Chem model simulates neutral (or weakly stable) layers below 1.5 km, which may limit 465 uplifting of aerosols from the surface, failing to create a deep well-mixed layer of aerosols (Fig. 466 12c). Although the dust emission at the surface is overestimated in AMJ in the 4km_D2 run, the 467 simulated neutral or weakly stable thermal structure does not favor convective vertical mixing, 468 resulting in the low biases of aerosols between 0.3 km and 3 km.

Similar biases of aerosol and instability in the lower troposphere are also shown in JAS (Fig. 12d and 13d). The stable boundary layer limits vertical transport of aerosols from the surface, contributing to the low bias of column-integrated AOD in JAS (Fig. 11). In JAS (Fig. 12d), aerosol extinction close to the CALIOP observation is simulated in the free atmosphere, suggesting that the low bias in AOD is not due to the halved chemical boundary conditions from MOZART-4. In the cold season, in spite of some discrepancies in the magnitude of atmospheric stability, all of the

simulations capture the stable lower troposphere (Fig. 13a and 13b), consistent with relativelygood performance of aerosol simulations in the cold season.

477 As biases in the model simulations are found mainly within the boundary layer, a sensitivity 478 experiment is conducted at 20 km resolution using the ACM2 PBL scheme (20km P7). Although 479 the changes in the meteorological variables (Supplementary Fig. 6-9) and atmospheric static 480 stability (Fig. 13) are rather small, the simulated surface NO_3 and NH_4 in the 20km_P7 run 481 decrease by 50% compared to the 20km D2 run (Fig. 14c, 14d and Table 3). Considering that 482 more NO_3 and NH_4 are simulated at 4 km resolution than at 20 km resolution as shown in section 483 4.1, the use of the ACM2 PBL scheme at 4 km simulation would largely resolve the high biases 484 of NO₃ and NH₄ in the 4km_D2 simulation. The decrease of NO₃ and NH₄ near the surface is 485 because more aerosols are transported to the layers above 0.5 km (Fig. 15a and 15b), possibly 486 resulting from different convective vertical mixing in the PBL schemes. However, PM_{25} dust is 487 significantly overestimated by a factor of 4 in the 20km_P7 simulation (Fig. 14h), leading to a 488 small decrease of PM_{2.5} by only 8% compared with the 20km_D2 run in the cold season. In the warm season, PM2.5_dust in the 20km_P7 run is overestimated by a factor of 5, causing an 489 490 overestimation of PM_{2.5} and PM₁₀ (Fig. 14a and 14b). Aerosol extinctions in the boundary layer 491 above the surface increase in the warm season (Fig. 15c and 15d), possibly related to 492 overestimation of dust emissions and more conducive convective vertical transport in the PBL 493 scheme.

In summary, the WRF-Chem model captures the seasonal variations of meteorological variables (temperature, RH, wind speed and precipitation), despite some deviations in magnitude. The low biases in aerosol optical properties of the warm season likely do not originate from hygroscopic effects, wet removal processes or dust emissions associated with the wind speed bias. 498 The model simulates a stable environment in the warm season, which is opposite to the observed 499 unstable environment. The simulated stable environment may be most likely responsible for low 500 biases in the aerosol extinction above the surface (0.3-3 km) and the column-integrated AOD in 501 the warm season. Switching to the ACM2 PBL scheme leads to improved vertical displacement of 502 aerosols in the boundary layer, thus an improvement in the simulations of NO_3 and NH_4 in the cold 503 season. However, dust emissions are significantly overestimated with the ACM2 PBL scheme, 504 which contributes partly to the better simulation of aerosol extinction in the boundary layer and 505 AOD in the column. These results highlight that improving the simulation of boundary layer 506 structure and processes are critical for capturing the vertical profiles of aerosol extinction.

507 **4.4 Results in Rural Areas**

In general, low values of PM concentration are observed in the rural areas, Pinnacles and Kaiser (Fig. 16 and 17). The rural areas share some similar model performance to the urban areas, such as the overestimation of NO_3 , reasonable simulation of EC, good representation of SO_4 in the cold season and underestimation of SO_4 in the warm season. However, the results are not sensitive to model resolution. It suggests that high resolution is particularly important for heavily polluted areas due to the inhomogeneity of emission sources, but less important for relatively lightly polluted areas.

In late July/early August, MODIS (Moderate Resolution Imaging Spectroradiometer) fire data (not shown) showed active wild fires close to Kaiser, which resulted in high concentration of aerosols locally (Fig. 17). Our model simulations with monthly-varying fire emissions fail to reproduce these fire events. Previous studies (e.g., Grell et al., 2011; Wu et al. 2011a; Archer-Nicholls et al., 2015) demonstrated that the WRF-Chem model can capture aerosols distributions from wild fires based on fire locations from satellite observations. Campbell et al. (2016) further 521 described the difficulties in constraining total aerosol mass from operational satellite fire 522 observations and the time needed by the model for diffusion within the near-surface layers to 523 render both reasonable AOD and vertical profiles of aerosol extinction. For operational application 524 of the WRF-Chem model in MAIA retrievals, the observations of daily fire events need to be more 525 appropriately considered.

526 **5. Summary**

527 The WRF-Chem (Weather Research and Forecasting model with Chemistry) model is 528 employed to simulate the seasonal variability of aerosols in WY2013 (water year 2013) in the SJV 529 (San Joaquin Valley). Model simulations are evaluated using satellite and in-situ observations. In 530 general, the model simulations conducted at 4 km resolution reproduce the spatial and temporal 531 variations of regional aerosols in the cold season, when aerosols are mainly contributed to by 532 anthropogenic emissions in the SJV. The magnitude of simulated aerosols in the cold season 533 however, especially in relatively dense urban areas, is sensitive to model horizontal resolution. 534 The 4km simulation has comparable magnitude to available observations, while the 20km 535 simulation underestimates aerosols. Differences in aerosol simulation fidelity as a function of 536 variable resolutions are mainly due to the difference in aerosol emissions and simulated 537 Emissions at higher resolution can better resolve the inhomogeneity precipitation. of 538 anthropogenic emissions in the SJV than at lower resolution. The sensitivity to horizontal resolution is small in rural areas and during warm season, where/when the relative contribution of 539 540 anthropogenic emissions is small.

541 Previous studies in the SJV are mainly focused on $PM_{2.5}$ (particulate matter with diameter 542 $\leq 2.5 \mu m$) and during cold season (e.g. Chow et al., 2006; Herner et al., 2006; Pun et al., 2009; 543 Ying and Kleeman, 2009; Zhang et al., 2010; Chen et al., 2014; Hasheminassab et al., 2014; Kelly

et al., 2014; Baker et al., 2015; Brown et al., 2016). CALIOP (Cloud-Aerosol Lidar with 544 545 Orthogonal Polarization) and IMPROVE (Interagency Monitoring of Protected Visual 546 Environments) observations show that dust is a primary contributor to the aerosols in the SJV, 547 especially in the warm season. Dust contributes 24.6% to $PM_{2.5}$ while more than 75.8% to PM_{10} in 548 the warm season. For all seasons, the major component of aerosol extinction in the boundary layer 549 is dust as observed by CALIOP, consistent with Kassianov et al. (2012). For a complete 550 understanding of aerosol impacts on air quality, weather and climate, the full spectrum of aerosols 551 should be considered during all seasons.

552 All the model simulations conducted fail to capture aerosol vertical distribution and 553 variability in the SJV warm season, largely due to the misrepresentation of dust emissions, static 554 stability and vertical mixing in the boundary layer. The GOCART (Goddard Global Ozone 555 Chemistry Aerosol Radiation and Transport) dust emission scheme significantly underestimates 556 dust due to the non-active source function, S, for potential wind erosion used in this study while 557 the DUSTRAN (DUST TRANsport model) scheme may overestimate dust emission in the SJV. 558 Along with the bias in dust emissions, our simulations produce a relatively stable boundary layer 559 in the warm season, in contrast with observations suggesting a more unstable environment, leading to a weak vertical mixing of aerosols in the boundary layer. Improved dust emission and better 560 561 simulations of the boundary layer properties are needed for accurate simulation of aerosols in the 562 SJV warm season.

563 Other biases are also identified in the model simulations. NO_3 and NH_4 in the cold season 564 are overestimated in the model, but the results are sensitive to the choice of the PBL (planetary 565 boundary layer) scheme. The SOA (secondary organic aerosol) processes contribute to the 566 underestimation of OM (organic matter) in this study. The underestimation of sulfate in the warm 567 season may be caused by the misrepresentation of emissions and the chemical boundary conditions 568 related to marine intrusions. Aerosols from wild fires are not captured in the simulations with 569 monthly updated fire data. Further investigations are needed to improve model simulations in the 570 SJV for both scientific and operational applications.

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867 List of Table

868 Table 1. Experiment description

Experiment ID	Experiment description
_	
20km	Simulation with the GOCART dust scheme at 20 km horizontal resolution.
20km_D2	Same as 20km, but with the DUSTRAN dust scheme.
20km_P7	Same as 20km_D2, but with the ACM2 PBL scheme.
4km	Same as 20km, but at 4 km horizontal resolution.
4km_D2	Same as 4km, but with the DUSTRAN dust scheme.

Species	20km	4km	4km_D2	20km_D2	20km_P7
PM _{2.5}	0.89	0.90	0.86	0.78	0.03
PM _{2.5} _NO ₃	0.94	0.95	0.94	0.94	0.91
PM _{2.5} _NH ₄	0.97	0.96	0.96	0.98	0.96
PM _{2.5} _OM	0.93	0.93	0.94	0.93	0.91
PM _{2.5} _EC	0.98	0.98	0.98	0.98	0.96
PM _{2.5} _SO ₄	0.63	-0.16	-0.14	0.61	0.63
PM _{2.5} _dust	-0.55	-0.50	0.48	0.55	0.36
PM ₁₀	-0.25	-0.23	-0.08	0.01	-0.03

870 Table 2. Correlation with observations for different species at Fresno, CA

Species	Cold season						Warm season					
Species							Warm Season					
	OBS	20km	4km	4km_	20km	20km	OBS	20km	4km	4km_	20km	20km
				D2	_D2	_P7				D2	_D2	_P7
PM _{2.5}	16.84	13.71	21.38	22.48	14.90	13.77	8.44	4.91	6.29	12.85	10.12	14.85
PM _{2.5} NO ₃	5.43	6.36	9.54	9.22	6.22	3.16	0.84	0.55	0.69	0.79	0.66	0.57
PM _{2.5} NH ₄	1.42	1.97	2.99	2.88	1.91	0.98	0.40	0.19	0.24	0.20	0.16	0.13
PM _{2.5} OM	5.39	0.92	2.07	2.07	0.93	1.04	2.47	0.49	0.87	0.87	0.50	0.55
PM _{2.5} EC	1.08	0.52	1.12	1.13	0.52	0.58	0.32	0.27	0.49	0.49	0.27	0.30
PM _{2.5} SO ₄	0.87	0.53	0.82	0.81	0.53	0.46	1.04	0.54	0.61	0.60	0.53	0.49
PM _{2.5} _ dust	0.90	0.11	0.11	1.65	1.50	4.18	2.08	0.04	0.03	6.49	5.16	10.05
PM10	31.55	14.93	22.81	28.32	20.10	24.52	34.82	7.08	8.69	38.12	30.19	48.02

872 Table 3. Surface aerosol mass ($\mu g m^{-3}$) for different species at Fresno, CA

	4km_D2	20km_D2	20km_P7
Т	0.94	0.94	0.94
RH	0.98	0.98	0.96
Wind	0.83	0.84	0.85
Rain	0.97	0.97	0.97

874 Supplementary Table 1. Correlation with surface observations for meteorological variables at
 875 Fresno, CA

		Cold season		Warm season			
	4km_D2	20km_D2	20km_P7	4km_D2	20km_D2	20km_P7	
T (K)	3.89	3.56	3.69	2.44	1.50	1.35	
RH (%)	-9.78	-14.55	-19.35	-9.48	-9.32	-11.16	
Wind (m/s)	-0.67	-1.00	-1.05	0.78	-0.16	-0.49	
Rain (mm/day)	-0.15	0.14	-0.03	-0.06	-0.03	-0.04	

877 Supplementary Table 2. Bias for surface meteorological variables at Fresno, CA

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Figure 1. Daily mean anthropogenic $PM_{2.5}$ emission rate (µg m⁻² hr⁻¹) at (a) 20km and (b) 4km simulation. Domain-averaged emission rate is shown at right corner of each figure. Red dashed lines in Figure 1a represent the region used for the domain averages in the discussions. Yellow circle: IMPROVE site; yellow diamond: EPA CSN site. Three urban sites: Fresno, Bakersfield and Modesto; two rural sites: Pinnacles and Kaiser.



Figure 2. (a) Monthly precipitation (mm/day) from CPC, 20km and 4km; (b) monthly wind speed (m/s) from CIMIS, 20km and 4km. 4km_D2 (not shown) is similar to 4km.



Figure 3. Spatial distribution of seasonal mean 550 nm AOD from MISR and the WRF-Chem
(20km and 4km) simulations in WY2013. OND: October-November-December; JFM: JanuaryFebruary-March; AMJ: April-May-June; JAS: July-August-September.



Figure 4. (a) Monthly mean 550 nm AOD; (b) monthly mean 400-600 nm Ångström exponent at
Fresno, CA from October 2012 to September 2013.



Figure 5. Aerosol mass (μ g m⁻³) for different species from OBS, the 20km and 4km simulations at Fresno, CA. NH₄ observations are from EPA; other observations are from IMPROVE. PM_{2.5}_NO₃ represents NO₃ with diameter $\leq 2.5 \mu$ m. Similar definition for NH₄, EC, OM and SO₄ in the figures.





902 Figure 6. (a) NO_2 and (b) SO_2 from EPA (OBS) and the 20km run at Fresno, CA.



904 Figure 7. (a) $PM_{2.5}$ dust; (b) $PM_{2.5}$; and (c) PM_{10} from IMPROVE, the 4km and 4km_D2 905 simulations at Fresno, CA.



907 Figure 8. Mean dust emission rate ($\mu g m^{-2} s^{-1}$) from the 4km and 4km_D2 runs.



909 Figure 9. Fraction of erodible surface in the GOCART dataset used in this study.



Figure 10. Relative contribution (%) of aerosol species from IMPROVE and the WRF-Chem (4km and 4km_D2) simulations at Fresno, CA in WY2013. (Panel 1) Contribution to $PM_{2.5}$ in the cold season; (Panel 2) relative contribution of $PM_{2.5}$ and coarse mass (CM) to PM_{10} in the cold season; (Panel 3) same as Panel 1 but in the warm season; (Panel 4) same as Panel 2 but in the warm season. "Other" refers to the difference of $PM_{2.5}$ total mass and specified $PM_{2.5}$ (NO₃, NH₄, OM, EC, SO₄ and dust).



918 Figure 11. Spatial distribution of seasonal mean 550 nm AOD from the 4km_D2 run in WY2013.



Figure 12. Vertical distribution of seasonal mean 532 nm aerosol extinction coefficient (km⁻¹)
from CALIOP (blue) and the WRF-Chem (4km and 4km_D2) simulations over the red box
region in Fig. 1a in WY2013. Blue dashed lines (CALIOP_nodust) represent the CALIOP
profiles without dust (dust and polluted dust).





Figure 13. Vertical distribution of season mean equivalent potential temperature (θ_e ; K) from AIRS, ERA-Interim (ERA-I) and the WRF-Chem (4km_D2, 20km_D2 and 20km_P7) simulations over the red box region in Fig. 1a in WY2013. The 4km run (not shown) is similar to the 4km_D2 run.



Figure 14. Aerosol mass (μ g m⁻³) for different species from OBS, the 4km_D2, 20km_D2 and 20km_P7 simulations at Fresno, CA. NH₄ observations are from EPA; other observations are from IMPROVE. PM_{2.5}_NO₃ represents NO₃ with diameter \leq 2.5 µm. Similar definition for NH₄, EC, OM, SO₄ and dust in the figures.



Figure 15. Vertical distribution of seasonal mean 532 nm aerosol extinction coefficient (km⁻¹)
from CALIOP, CALIOP_nodust, and the WRF-Chem (4km_D2, 20km_D2 and 20km_P7)
simulations over the red box region in Fig. 1a in WY2013.



Figure 16. Aerosol mass (µg m⁻³) for different species from IMPROVE (OBS), the 4km_D2,
20km_D2 and 20km_P7 simulations at Pinnacles, CA.



Figure 17. Aerosol mass (µg m⁻³) for different species from IMPROVE (OBS), the 4km_D2,
20km_D2 and 20km_P7 simulations at Kaiser, CA.



Supplementary Figure 1. Spatial distribution of seasonal mean 550 nm AOD from MISR and the
WRF-Chem (20km_D2, 20km_P7, 20km_BC1 and 20km_NEI11) simulations in WY2013. OND:
October-November-December; JFM: January-February-March; AMJ: April-May-June; JAS: JulyAugust-September. The 20km_BC1 run is the same as the 20km_D2 run except that chemical
boundary conditions use MOZART-4 original data. The 20km_NEI11 run is the same as the
20km_D2 run except with NEI11 anthropogenic emissions.



Supplementary Figure 2. Aerosol mass ($\mu g \text{ m}^{-3}$) for different species from OBS, the 20km_D2, 20km_BC1 and 20km_NEI11 simulations at Fresno, CA. NH₄ observations are from EPA; other observations are from IMPROVE. PM_{2.5}_NO₃ represents NO₃ with diameter \leq 2.5 μm . Similar definition for NH₄, EC, OM, SO₄ and dust in the figures.



Supplementary Figure 3. Vertical distribution of seasonal mean 532 nm aerosol extinction
coefficient (km⁻¹) from CALIOP, CALIOP_nodust, and the WRF-Chem (20km_D2, 20km_BC1
and 20km_NEI11) simulations over the red box region in Fig. 1a in WY2013.



Supplementary Figure 4. Aerosol mass ($\mu g m^{-3}$) for different species from EPA CSN (OBS), the 4km_D2, 20km_D2 and 20km_P7 simulations at Bakersfield, CA. PM_{2.5}_NO₃ represents NO₃ with diameter $\leq 2.5 \mu m$. Similar definition for SO₄, EC, OM, NH₄ and dust in the figures.



Supplementary Figure 5. Aerosol mass (µg m⁻³) for different species from EPA CSN (OBS), the
4km_D2, 20km_D2 and 20km_P7 simulations at Modesto, CA.





967 Supplementary Figure 6. Monthly mean of (a) 2-m temperature (°C); (b) 2-m relative humidity 968 (%); (c) 10-m wind speed (m/s); (d) precipitation (mm/day) at Fresno, CA. The 20km (not shown) 969 run is similar to the 20km_D2 run while the 4km (not shown) run is similar to the 4km_D2 run.



Supplementary Figure 7. Vertical profile of seasonal mean temperature (K) bias in the WRF-Chem
simulations and AIRS comparing to ERA-Interim. The 20km run (not shown) is similar to the
20km_D2 run while the 4km run (not shown) is similar to the 4km_D2 run.



Supplementary Figure 8. Vertical profile of seasonal mean relative humidity (%) in the WRF-Chem
simulations, AIRS and ERA-Interim. The 20km run (not shown) is similar to the 20km_D2 run
while the 4km run (not shown) is similar to the 4km D2 run.



Supplementary Figure 9. Vertical profile of seasonal mean specific humidity (g kg⁻¹) in the WRFChem simulations, AIRS and ERA-Interim. The 20km run (not shown) is similar to the 20km_D2
run while the 4km run (not shown) is similar to the 4km D2 run.