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1 **WRF-Chem simulation of aerosol seasonal** 2 **variability in the San Joaquin Valley**

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18 Highlights:

- 19 1. The WRF-Chem simulation successfully captures aerosol variations in cold season in the San
20 Joaquin Valley (SJV), but has poor performance in 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 cold
23 season in the SJV.
- 24 3. Observations show that dust is a major component of aerosols in the SJV, especially in warm
25 season. Poor performance of the WRF-Chem model in warm season in the SJV is mainly due
26 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 in cold
31 season. However, the aerosols are not well represented in warm season. Aerosol simulations in
32 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
34 distribution of anthropogenic emissions. In rural areas, the model sensitivity to grid size is rather
35 small. Our observational analysis show that dust is a primary contributor to aerosols in the SJV,
36 especially in the warm season. Aerosol simulations in the warm season are sensitive to
37 parameterization of dust emission in the WRF-Chem model. The GOCART (Goddard Global
38 Ozone Chemistry Aerosol Radiation and Transport) dust scheme produces very little dust in the
39 SJV while the DUSTRAN (DUST TRANsport model) scheme overestimates dust emission.
40 Vertical mixing of aerosols is not adequately represented in the model comparing to CALIPSO
41 (Cloud-Aerosol Lidar and Infrared pathfinder Satellite Observation) aerosol extinction profiles.
42 Improved representation of dust emission and vertical mixing are needed for better simulations
43 of aerosols in warm season in the SJV. Aerosols generated by wild fires are not captured in the
44 simulations with climatological fire emissions, underscoring the need of fire emission
45 observations for operational usage.

46



47 **1. Introduction**

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

61 Air quality models are a critical tool to understand the formation and evolution of
62 aerosols and their impacts on air quality and climate. However, it is still quite a challenge to
63 accurately simulate aerosol properties (Fast et al., 2014). Fast et al. (2014) summarized the
64 factors contributing to the errors in region-scale modeling of aerosol properties, including 1)
65 emission sources; 2) meteorological parameterizations; 3) representation of aerosol chemistry; 4)
66 limited understanding of the formation processes of secondary organic aerosol (SOA); 5) spatial
67 resolution; and 6) boundary conditions.

68 As one of the advanced regional air quality models, the Weather Research and
69 Forecasting model with Chemistry (WRF-Chem) has been widely used to study aerosols and



70 their impacts on regional air quality and climate (e.g., Misenis and Zhang, 2010; Zhang et al.,
71 2010; Zhao et al., 2010; 2013; 2014; Wu et al., 2011a, 2011b, 2013; Fast et al., 2012, 2014;
72 Scarino et al., 2014; Tessum et al., 2015; Campbell et al., 2016; Hu et al., 2016). Fast et al.
73 (2014) showed that WRF-Chem simulations at 4 km horizontal resolution captured the observed
74 meteorology and boundary layer structure over California in May and June of 2010. The model
75 reasonably simulated the spatial and temporal variation of aerosols. Aerosol simulations by
76 WRF-Chem are usually sensitive to both local emission and long-range transport of aerosols
77 from the boundary conditions provided by the global Model for Ozone and Related chemical
78 Tracers, version 4 (MOZART-4). Similarly, in a one-year simulation at 12 km horizontal
79 resolution, Zhao et al. (2013) showed that the WRF-Chem model represented the observed
80 seasonal and spatial variation of surface particulate matter (PM) concentration over California.
81 However, underestimation of elemental carbon (EC) and organic matter were noticed in the
82 model simulation, with no sensitivity to horizontal model resolution.

83 In this study, we extend the studies by Fast et al. (2014) and Zhao et al. (2013) by
84 focusing on simulating aerosol seasonal variability in the most polluted SJV in California. This
85 paper serves as the first step for future investigation of the aerosol impact on regional climate
86 and the water cycle in California. Previous studies have demonstrated that aerosols are better
87 simulated at higher model resolution (Misenis and Zhang et al., 2010; Qian et al., 2010; Stround
88 et al., 2011; Fountoukis et al., 2013). However, most regional climate studies are still limited to
89 coarse model resolutions (on the order of 10 km) due to the availability of computational
90 resources. This study will investigate the sensitivity of aerosol simulations to horizontal
91 resolution and identify suitable model resolution for regional climate study in the SJV.



92 Another application of air quality modeling is to provide initial *a priori* input 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
95 Administration) MAIA (Multi-Angle Imager for Aerosols) mission, which aims to map PM
96 component concentrations in major urban areas (including the SJV). A reasonable initial estimate
97 of aerosol speciation from WRF-Chem is critical to ensure the retrieval speed and quality.
98 Considering the sensitivity of WRF-Chem simulations to various factors such as initial and
99 boundary conditions, model parameterizations and emission sources (e.g., Wu and Petty, 2010;
100 Zhao et al., 2010, 2013; Wu et al., 2011a, 2015; Fast et al., 2014; Campbell et al., 2016;
101 Morabito et al., 2016), careful model evaluations are needed before the simulations can be used
102 for remote sensing retrievals. This study also serves as an evaluation for WRF-Chem aerosol
103 simulations in the SJV, which will provide important information for utilizing WRF-Chem for
104 MAIA retrieval algorithms, critical to the success of the MAIA mission.

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

109 **2. Observations**

110 **2.1 Aerosol Optical Depth**

111 Aerosol optical depth (AOD) is a measure of column-integrated light extinction by
112 aerosols and a proxy for total aerosol loading in the atmospheric column. The Aerosol Robotic
113 Network (AERONET) provides ground measurements of AOD every 15 minutes during daytime
114 (Holben et al., 1998), with an accuracy of ± 0.01 (Eck et al., 1999; Holben et al., 2001). The



115 monthly level 2.0 product with cloud screening and quality control is used in this study.
116 AERONET AOD is interpolated to $0.55\mu\text{m}$ using the Ångström exponent. In the SJV, only one
117 AERONET station at Fresno, CA has regular observations throughout the California water year
118 2013 (WY2013; i.e., from October 2012 to September 2013).

119 The Multiangle Imaging Spectroradiometer (MISR) (Diner et al., 1998) instrument
120 onboard the Terra satellite has provided global coverage of AOD once a week since December
121 1999. The standard MISR retrieval algorithm provides AOD observations at 17.6 km resolution
122 using 16 x 16 pixels of 1.1 km each. About 70% of MISR AOD retrievals are within 20% of the
123 paired AERONET AOD, and about 50% of MISR AOD falls within 10% of the AERONET
124 AOD, except in the dusty and hybrid (smoke+dust) sites (Kahn et al., 2010). We use version 22
125 of Level 3 monthly AOD product at 0.5° resolution in this study.

126 **2.2 Surface Mass Concentration**

127 Surface PM_{2.5} speciation and PM₁₀ (particulate matter with diameter $\leq 10\ \mu\text{m}$) data are
128 routinely collected by two national chemical speciation monitoring networks: Interagency
129 Monitoring of Protected Visual Environments (IMPROVE) and the PM_{2.5} National Chemical
130 Speciation Network (CSN) operated by Environmental Protection Agency (EPA) (Hand et al.
131 2011; Solomon et al., 2014). IMPROVE collects 24-h aerosol speciation every third day at
132 mostly rural sites since 1988. The same frequency of aerosol speciation data was collected at
133 EPA CSN sites in urban and suburban areas since 2000. Selected IMPROVE and EPA CSN sites
134 used in this study are shown in Figure 1a.

135 **2.3 Aerosol Extinction Profile**

136 The aerosol extinction coefficient profile reflects the attenuation of the light passing
137 through the atmosphere due to the scattering and absorption by aerosol particles as a function of



138 range. Version 3 Level 2 532 nm aerosol extinction profiles derived from Cloud-Aerosol Lidar
139 with Orthogonal Polarization (CALIOP) backscatter profiles collected onboard the Cloud-
140 Aerosol Lidar and Infrared pathfinder Satellite Observation (CALIPSO) satellite are used (Omar
141 et al., 2009; Young and Vaughan, 2009). Seasonal mean profiles are derived for WY2013 based
142 on the methodology outlined in Campbell et al. (2012), whereby quality-assurance protocols are
143 applied to individual profiles before aggregating and averaging the data. We highlight that no
144 individual profiles are included in the averages if the CALIOP Level 2 retrieval failed to resolve
145 any extinction within the column, a potential biasing issue that has recently been described by
146 Toth et al. (2016). Level 2 532 nm aerosol extinction is speciated, with algorithms resolving
147 aerosol type present for clean marine, dust, polluted continental, clean continental, polluted dust
148 and smoke. Dust and polluted dust are specifically distinguished in the averages applied below
149 for their contribution to total extinction and the vertical profile seasonally in the SJV.

150 **2.4 Equivalent Potential Temperature**

151 Equivalent potential temperature (θ_e) is a quantity relevant to the stability of the air. The
152 θ_e profiles used in this study are derived from temperature and moisture profiles observed by AIRS
153 (Atmospheric Infrared Sounder) onboard the Aqua satellite (Susskind et al., 2003; Divakarla et al.,
154 2006). AIRS has provided global coverage of the tropospheric atmosphere at approximately 01:30
155 and 13:30 local time since 2002. AIRS retrievals have root-mean-squared (RMS) difference of ~1
156 K for temperature and ~15% for water vapor (Divakarla et al., 2006). Level 3 monthly temperature
157 and moisture retrievals (version 6) at $1^\circ \times 1^\circ$ grid are used in this study.

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

159 The WRF-Chem model Version 3.5.1 (Grell et al., 2005) updated by Pacific Northwest
160 National Laboratory (PNNL) is used in this study (Zhao et al., 2014). Similar to the chemical



161 parameterizations used in the Zhao et al. (2014), this study uses the CBM-Z (carbon bond
162 mechanism) photochemical mechanism coupled with the four-sectional-bin MOSAIC (Model for
163 Simulating Aerosol Interactions and Chemistry) aerosol scheme as the chemical driver. The major
164 components of aerosols (nitrate, ammonium, EC, organic carbon, sulfate, sea salt, dust, etc.) as
165 well as their physical and chemical processes are simulated in the model. More details of the
166 chemical settings used in this study can be found in Zhao et al. (2014) and references therein.

167 The model simulations start on 1 September 2012 and run continuously for 13 months.
168 With the first month as spin-up, our analysis focuses on WY2013 from October 2012 to September
169 2013. The model is configured with 40 vertical levels and a model top at 50 hPa. The model center
170 is placed at 38°N, 121°W, with 250 x 350 grids at 4 km horizontal resolution (referred to as “4km”
171 hereafter; Table 1), covering California and the surrounding area. To test the sensitivity of aerosol
172 simulations on horizontal resolution, one simulation with the same model settings and domain
173 coverage is conducted at 20 km horizontal resolution (referred to as “20km” hereafter).

174 The physics parameterizations used in the simulations include the Morrison double-
175 moment microphysics scheme (Morrison et al., 2009), Rapid Radiative Transfer Model for General
176 circulation model (RRTMG) shortwave and longwave radiation schemes (Iacono et al., 2008),
177 Yonsei University (YSU) planetary boundary layer scheme (Hong et al., 2006), Community Land
178 Model (CLM) Version 4 land surface scheme (Lawrence et al., 2011). Grell 3D ensemble cumulus
179 scheme (Grell and Devenyi, 2002) is used in the 20km simulation while the 4km simulation does
180 not use cumulus parameterization. The ERA-Interim reanalysis data (Dee et al., 2011) provides
181 meteorological initial and boundary conditions for the WRF-Chem. The MOZART-4 global
182 chemical transport model (Emmons et al., 2010) is used for the chemical initial and boundary
183 conditions. Fast et al. (2014) found that the MOZART-4 model has overestimation of aerosols in



184 the free troposphere over California. Following Fast et al. (2014), the chemical initial and boundary
185 conditions from MOZART-4 are divided by two in all simulations.

186 Anthropogenic emissions are provided by US EPA 2005 National Emissions Inventory
187 (NEI05), with area-type emissions on a structured 4-km grid and point type emissions at latitude
188 and longitude locations (US EPA, 2010). Anthropogenic emissions are updated every hour to
189 account for diurnal variability, while its seasonal variation is not considered in the simulations.
190 Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols
191 from Nature (MEGAN) model (Guenther et al., 2006). Biomass burning emissions are obtained
192 from the Global Fire Emissions Database, version 2.1 with eight-day temporal resolution
193 (Randerson et al., 2007). Sea salt emissions use the PNNL-updated sea salt emission scheme that
194 includes the correction of particles with radius less than $0.2 \mu\text{m}$ (Gong et al., 2003) and dependence
195 on sea surface temperature (Jaeglé et al., 2011).

196 Following Zhao et al. (2013), dust emission is computed from the GOCART (Goddard
197 Global Ozone Chemistry Aerosol Radiation and Transport) dust scheme (Ginoux et al., 2001) in
198 the 20km and 4km simulations. As shown later, a significant amount of dust is observed in the
199 SJV while the GOCART dust scheme produces little dust. One sensitivity experiment at 4 km
200 horizontal resolution (referred to as “4km_D2” hereafter) is conducted by switching dust emission
201 scheme to the DUST TRANsport model (DUSTRAN) scheme (Shaw et al., 2008). Detailed
202 descriptions of the two dust emission schemes can be found in Zhao et al. (2010).

203 **4. Model Simulation Results**

204 WRF-Chem model simulation results and their evaluations are in this section. We start the
205 discussions with a focus on the polluted urban areas. Because aerosols properties and model
206 performance are similar at all urban sites, our discussion is focused on the results at Fresno, CA



207 while those at other urban sites are provided in supplementary materials. Model simulations in
208 rural areas are presented in the last subsection.

209 **4.1 Sensitivity to Horizontal Resolution**

210 Figure 1 shows daily mean anthropogenic PM_{2.5} emission rates used in the 20km and 4km
211 simulations, respectively. Although both of the PM_{2.5} emission rates are derived from the 4 km
212 NEI05 dataset, localized high emission rates with sharp gradients are evident at urban areas in the
213 4km simulation (Figure 1b). The 20km simulation has lower emission rates with smoother features
214 due to the averaging process (Figure 1a).

215 Consistent with the emission rate differences, higher AOD is simulated at 4km than 20km,
216 mainly in cold season (OND and JFM in Figure 2). The 4km simulation reproduces the distribution
217 and magnitude of AOD observed by MISR well in the cold season. The AOD difference between
218 20km and 4km is small in the warm season (AMJ and JAS in Figure 2). Both the 20km and 4km
219 runs underestimate AOD in the warm season compared with MISR. Model performance identified
220 in Figure 2, including the sensitivity to horizontal resolution in cold season and underestimation
221 of AOD in warm season, are further confirmed by comparing to AERONET observations at Fresno,
222 CA (Figure 3). In cold season at Fresno, the AOD in the 20km simulation is 23% lower than the
223 AOD in the 4km simulation. The different model sensitivities to horizontal resolution from the
224 cold to the warm season suggest that the dominant aerosol sources are different through the two
225 seasons. We will elaborate upon the aerosol composition in the following section. AERONET
226 shows small seasonal variation of AOD in the SJV, which is not well represented in the 20km and
227 4km simulations (Figure 2 and 3).

228 Aside from AOD, significant seasonal variability of PM_{2.5} is observed in the SJV urban
229 areas (Figure 4a and Supplementary Figure 1a and 2a). PM_{2.5} at Fresno peaks in January (26.18



230 $\mu\text{g m}^{-3}$) and has minimum of $7.03 \mu\text{g m}^{-3}$ in June, with an annual nonattainment value of $12.64 \mu\text{g}$
231 m^{-3} in total (Figure 4a). All WRF-Chem simulations successfully capture the seasonal variability
232 of PM_{2.5} observed in the SJV.

233 In the cold season, the 4km simulation overestimates PM_{2.5} by 27% while the 20km
234 simulation exhibits a low bias of 19% compared with IMPROVE observations at Fresno (Table 2).
235 High PM_{2.5} concentrations are primarily nitrate. Both simulations produce seasonal variability of
236 nitrate, but with high biases of 17% in 20km and 75% in 4km in the cold season (Figure 4c). It
237 suggests that the NEI05 dataset may have a high bias in nitrate emissions, which was also found
238 in Texas (Kim et al., 2011). OC, the second largest contributor of cold season PM_{2.5} in the SJV,
239 is significantly underestimated by 76% in the 20km simulation (Figure 4f). The 4km simulation
240 produces more OC than the 20km simulation, but it is still lower than IMPROVE by 46%. Fast et
241 al. (2014) suggested that the low bias in the WRF-Chem simulation is primarily due to incomplete
242 understanding of SOA processes.

243 Significant underestimation of EC and sulfate in the cold season are also shown in the
244 20km simulation, while the 4km simulation exhibits good agreement with IMPROVE (Figure 4d
245 and 4e). Sulfate in both simulations exhibits a low bias of ~45% in the warm season. Low bias of
246 simulated sulfate, with a failure of capturing the peaks during late afternoon, was also shown at
247 Bakersfield in Fast et al. (2014). It suggests that improvement in understanding the photochemical
248 processes involving sulfate is needed to reproduce seasonal variability of sulfate in the SJV. The
249 4km simulation of PM₁₀ has good agreement with IMPROVE in winter (December, January and
250 February), but a large low bias is found in other months (Figure 4b). The 20km simulation
251 underestimates PM₁₀ throughout WY2013.



252 Overall, the 4km simulation produce higher AOD and surface PM than the 20km
253 simulation in urban areas of the SJV, especially in the cold season. The 4km simulation has better
254 agreement with satellite and surface observations than the 20km simulation. The 4km simulation
255 captures seasonal variability of PM_{2.5} and its speciation. However, significant underestimation of
256 AOD and PM₁₀ are shown during the warm season in both 4km and 20 km simulations. The
257 underestimation also exists in a sensitivity experiment initialized in April (not shown). The
258 relatively good performance in simulating PM_{2.5} but PM₁₀ suggests that coarse aerosol particle
259 mass (CM; 10 μm ≥ particulate matter with diameter > 2.5 μm), mainly dust in the SJV, is not
260 represented well in the simulations. The impact of dust parameterizations is investigated in the
261 4km_D2 experiment.

262 4.2 Sensitivity to Dust Scheme

263 Limited amounts of PM_{2.5_dust} (dust with diameter ≤ 2.5 μm) are observed in the SJV
264 cold season, with a minimum in December (Figure 5c). The amount of PM_{2.5_dust} increases in
265 the warm season, with a peak in September. The 4km simulation produces comparable PM_{2.5_dust}
266 to IMPROVE in the winter, but almost no dust in other months. The 4km_D2 simulation represents
267 well the magnitude of PM_{2.5_dust} in cold season. However, too much PM_{2.5_dust} is simulated
268 in warm season, resulting in an overestimation of PM_{2.5} by 52% (Figure 5b and Table 2). Both
269 the 4km and 4km_D2 simulations capture seasonal variability of PM_{2.5}, but not for PM₁₀ (Figure
270 5a). The magnitude of PM₁₀ in the 4km_D2 is larger than the 4km simulation. PM₁₀ in the
271 4km_D2 is overestimated in AMJ but underestimated in JAS, leading to comparable season mean
272 with IMPROVE observations.

273 On the relative contribution of different aerosol species, IMPROVE observations at Fresno
274 show that nitrate is the primary contributor (32.3%) to PM_{2.5} while only 5.3% of PM_{2.5} is dust



275 in the cold season (panel 1 of Figure 6). Both 4km and 4km_D2 roughly reproduce the relative
276 contributions to PM_{2.5} in the cold season, with an overestimation of nitrate and underestimation
277 of OC found in Figure 4. Relative contributions of dust to PM_{2.5} are better simulated in 4km_D2
278 than in 4km. IMPROVE shows that 46.6% of PM₁₀ is in the cold season (panel 2 of Figure 6).
279 Both 4km (6.3%) and 4km_D2 (20.6%) underestimate the contribution of CM to PM₁₀. In the
280 warm season, dust (24.6%) becomes the primary contributor to PM_{2.5} while the contribution from
281 nitrate decreases to 9.9% as observed by IMPROVE (panel 3 of Figure 6). Almost no PM_{2.5_dust}
282 is simulated in 4km while too much PM_{2.5_dust} is produced in 4km_D2 in the warm season. The
283 relative contribution of CM to PM₁₀ is too small (27.6%) in 4km while 4km_D2 has better relative
284 contribution of 66.3% comparing to IMPROVE observed 75.8% (panel 4 of Figure 6).

285 AOD simulations are improved in the 4km_D2 experiment (Figure 7), with better
286 agreement with MISR (Figure 2). AOD in 4km_D2 is comparable to observations in AMJ, but still
287 underestimated in JAS. Consistent with AOD, the vertical distribution of aerosol extinction is
288 reasonably simulated in cold season in the WRF-Chem simulations while large discrepancies are
289 shown in warm season (Figure 8). As observed by CALIOP at 532 nm, aerosols are mainly
290 confined below 1 km above the surface in the cold season. Model simulations reasonably capture
291 the vertical distribution of aerosol extinction observed by CALIOP, with low biases in the
292 boundary layer and high biases in the free atmosphere. Similar discrepancy between the model
293 simulations and CALIOP is shown in other studies (Wu et al., 2011a; Hu et al., 2016). The
294 difference between 4km and 4km_D2 is small in cold season.

295 Dust in the boundary layer is a primary factor contributing to aerosol extinction in the SJV,
296 as illustrated by the differences between the bulk seasonal CALIOP mean profile and those
297 excluding the contributions of the dust and polluted dust species (CALIOP_nodust) profiles



298 (Figure 8). The simulated aerosol extinctions fall between the two in all seasons, suggesting
299 relatively good performance of simulating aerosols except for dust. Although a small portion of
300 PM_{2.5} is dust in the cold season, dust contributes to about 50% of total aerosol extinction (Figure
301 8a and 8b). A predominate portion of aerosol extinction in the boundary layer is contributed to by
302 dust in the warm season (Figure 8c and 8d). There, the 4km_D2 simulation produces higher aerosol
303 extinction in the boundary layer than the 4km simulation, though it is still lower than CALIOP.

304 Overall, poor simulations of dust play the dominant role in the bias of aerosols, especially
305 in warm season. Both the GOCART and DUSTRAN dust emission schemes used in this study
306 have problems in reproducing dust emission in the SJV, with underestimation in GOCART and
307 overestimation in DUSTRAN (Figure 5c). Improvement on dust emission is required for correctly
308 simulating seasonal variability of aerosols in the SJV.

309 **4.3 The Role of Meteorology**

310 In the warm season, more aerosols are observed at higher altitude than during the cold
311 season (Figure 8). A well-mixed layer of aerosols is observed below 1.5 km in AMJ (Figure 8c),
312 consistent with the large instability below 1.5 km observed by AIRS (Figure 9c). Both simulations
313 fail to capture this mixed layer of aerosols (Figure 8c) due to weak vertical mixing as evidenced
314 by relatively small instability in the simulations (Figure 9c). Aerosol extinction gradually
315 decreases with height in the simulations (Figure 8c). Similar biases of aerosol and instability in the
316 boundary layer are also shown in JAS (Figure 8d and 9d). Weak instability in the simulation, which
317 limits vertical mixing of aerosols, likely enhances the low bias of JAS AOD (Figure 7). Although
318 the 4km_D2 experiment produces comparable AOD and surface mass in AMJ (Figure 5 and Figure
319 7), the vertical distribution of aerosols is not well represented (Figure 8). The comparable AOD in
320 4km_D2 results from the low bias in the boundary layer and high bias in the free atmosphere. The



321 high bias in the free atmosphere suggests that the low bias in AOD are not due to the halved
322 chemical boundary conditions from MOZART-4. The stability biases in cold season are relatively
323 small (Figure 9a and 9b), consistent with good performance of aerosol simulation in the cold
324 season. These results highlight that the vertical mixing of dust must be correctly represented in
325 order to resolve the aerosol extinction profile correctly. Improved simulation of boundary layer
326 physics and dynamics during the warm season in the SJV warrants future investigation.

327 **4.4 Results in Rural Areas**

328 In general, low values of PM concentration are observed in the rural areas, Pinnacles and
329 Kaiser (Figure 10 and 11). The rural areas share some similar model performance with the urban
330 areas, such as the overestimation of nitrate, reasonable simulation of EC, good representation of
331 sulfate in cold season and underestimation of sulfate in warm season. However, the sensitivity to
332 model resolution is not significant. It suggests that high model resolution is particularly important
333 for heavily polluted areas due to the inhomogeneity of emission sources, but less important for
334 relatively lightly polluted areas.

335 In late July/early August, MODIS (Moderate Resolution Imaging Spectroradiometer) fire
336 data (not shown) observed active wild fires close to Kaiser, which resulted in high concentration
337 of aerosols at Kaiser (Figure 11). Our model simulations with climatological fire emissions fail to
338 reproduce these fire events. Based on fire locations from satellite observations, Wu et al. (2011a)
339 has demonstrated that the WRF-Chem model can capture aerosols distributions from wild fires
340 over South America. Campbell et al. (2016) further described the difficulties in both constraining
341 total aerosol mass from operational satellite fire observations and the time necessary within the
342 model for diffusion within the near-surface layers to render both reasonable AOD and vertical



343 profiles of aerosol extinction. For operational application of the WRF-Chem model in MAIA
344 retrievals, the observations of fire events need to be considered.

345 **5. Summary**

346 The WRF-Chem model is applied to simulate seasonal variability of aerosols in WY2013
347 (water year 2013) in the SJV (San Joaquin Valley). Model simulations are evaluated using satellite
348 and in-situ observations. In general, the model simulations at 4 km resolution reproduce the spatial
349 and temporal variations of aerosols in cold season, when aerosols are mainly contributed by
350 anthropogenic emissions in the SJV. The magnitude of simulated aerosols in the cold season,
351 especially in the urban areas, is sensitive to model horizontal resolution. The 4km simulation has
352 comparable magnitude to the observations while the 20km simulation underestimates aerosols.
353 The differences of aerosol simulations between different model resolutions are mainly due to the
354 difference in aerosol emissions. Emissions at higher resolution can better resolve the
355 inhomogeneity of anthropogenic emissions in the SJV than at lower resolution. The sensitivity to
356 horizontal resolution is small in the rural areas and in warm season, when the contribution of
357 anthropogenic emissions is small.

358 Previous studies in the SJV are mainly focused on PM_{2.5} (particulate matter with diameter
359 $\leq 2.5 \mu\text{m}$) and during cold season (e.g. Chow et al., 2006; Herner et al., 2006; Pun et al., 2009;
360 Ying and Kleeman, 2009; Zhang et al., 2010; Chen et al., 2014; Hasheminassab et al., 2014; Kelly
361 et al., 2014; Baker et al., 2015; Brown et al., 2016). CALIOP (Cloud-Aerosol Lidar with
362 Orthogonal Polarization) and IMPROVE (Interagency Monitoring of Protected Visual
363 Environments) observations show that dust is a primary contributor to aerosols in the SJV in warm
364 season. Dust contributes 24.6% to PM_{2.5} while more than 75.8% to PM₁₀ (particulate matter with
365 diameter $\leq 10 \mu\text{m}$) in warm season. For all seasons, the major component of aerosol extinction in



366 the boundary layer is dust as observed by CALIOP, consistent with Kassianov et al. (2012). For a
367 complete understanding of aerosol impact on air quality and regional climate, the full spectrum of
368 aerosols should be considered during all seasons.

369 All the model simulations fail to capture aerosol distribution and variability in the SJV
370 warm season, largely due to the misrepresentation of dust emission and vertical mixing. The
371 GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport) dust emission
372 scheme significant underestimates dust while the DUSTRAN (DUST TRANsport model) scheme
373 may overestimate dust emission in the SJV. Along with the bias in dust emissions, our simulations
374 produce weak atmospheric instability in warm season, leading to weak vertical mixing. Improved
375 dust emission and better simulations of boundary layer properties are needed for correct simulation
376 of aerosols in warm season in the SJV.

377 Other biases are also identified in the model simulations. Nitrate in the cold season is
378 overestimated in the model, possibly due to the overestimation of emissions. Incomplete
379 understanding of SOA (secondary organic aerosol) could contribute to the underestimation of OC
380 (organic carbon). Underestimation of sulfate in the warm season may be due to incorrect
381 photochemical processes of sulfate in the model. Aerosols from wild fires are not captured in the
382 simulations with climatological fire emissions. Further investigations are needed to improve model
383 simulations in the SJV for both scientific and operational applications. The evaluation framework
384 used in this study can be used to other polluted regions to ensure that aerosols are simulated
385 correctly for the right reasons.

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610 2014.



611 **List of Table**

612 Table 1. Experiment description

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

613

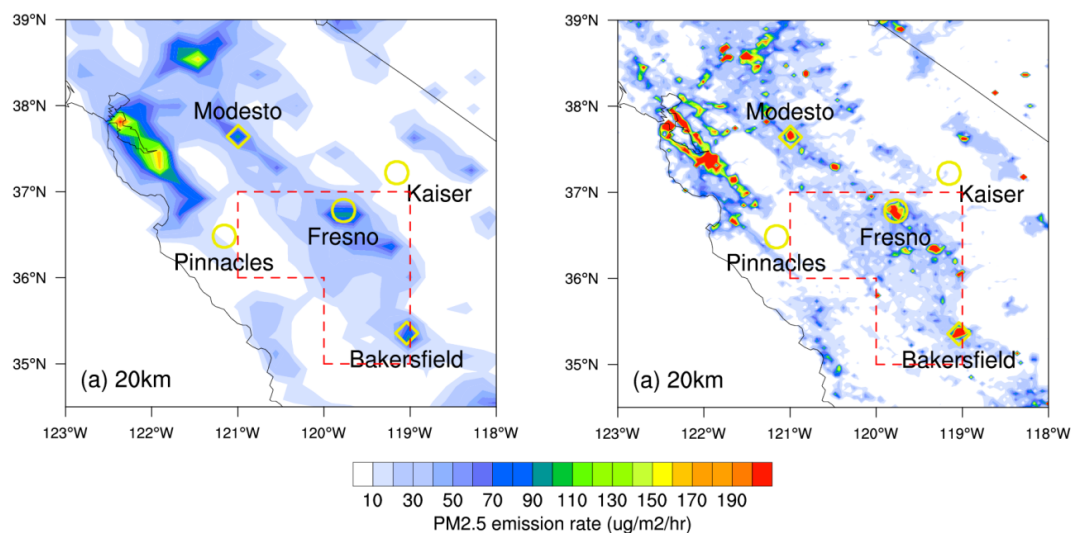
614 Table 2. Surface aerosol mass ($\mu\text{g m}^{-3}$) for different species at Fresno, CA

Species	Cold season				Warm season			
	IMPROVE	20km	4km	4km_D2	IMPROVE	20km	4km	4km_D2
PM2.5	16.84	13.71	21.38	22.48	8.44	4.91	6.29	12.85
PM2.5_NO ₃	5.43	6.36	9.54	9.22	0.84	0.55	0.69	0.79
PM2.5_OC	3.85	0.92	2.07	2.07	1.76	0.49	0.87	0.87
PM2.5_EC	1.08	0.52	1.12	1.13	0.32	0.27	0.49	0.49
PM2.5_SO ₄	0.87	0.53	0.82	0.81	1.04	0.54	0.61	0.60
PM2.5_dust	0.90	0.11	0.11	1.65	2.08	0.04	0.03	6.49
PM10	31.55	14.93	22.81	28.32	34.82	7.08	8.69	38.12

615

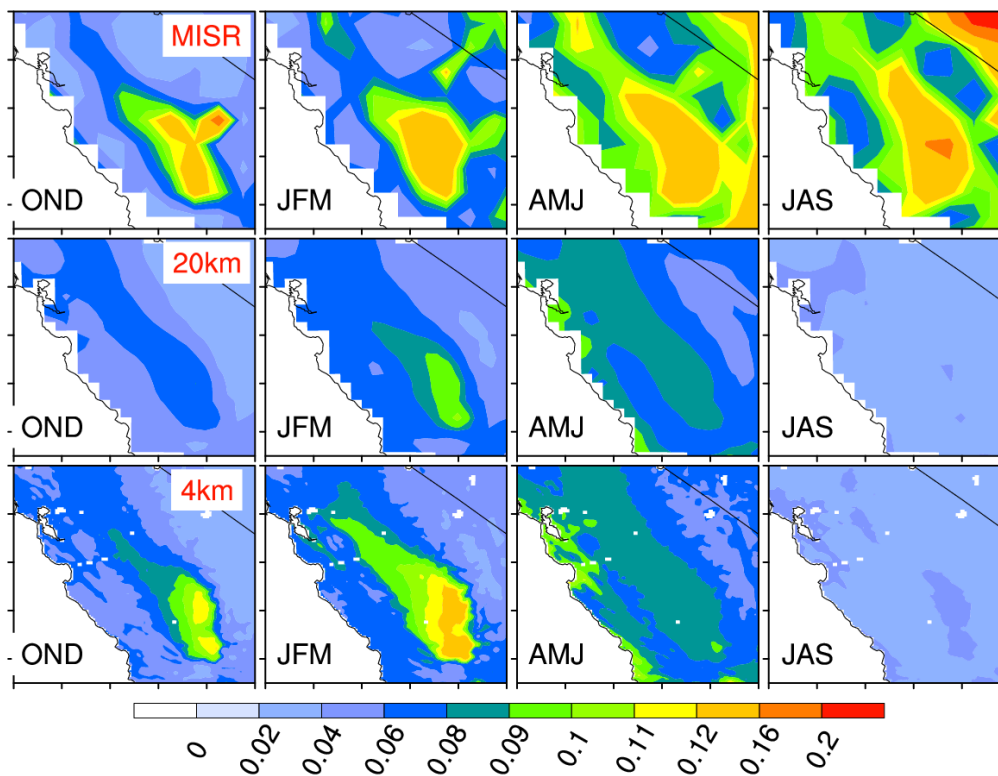


616 List of Figures



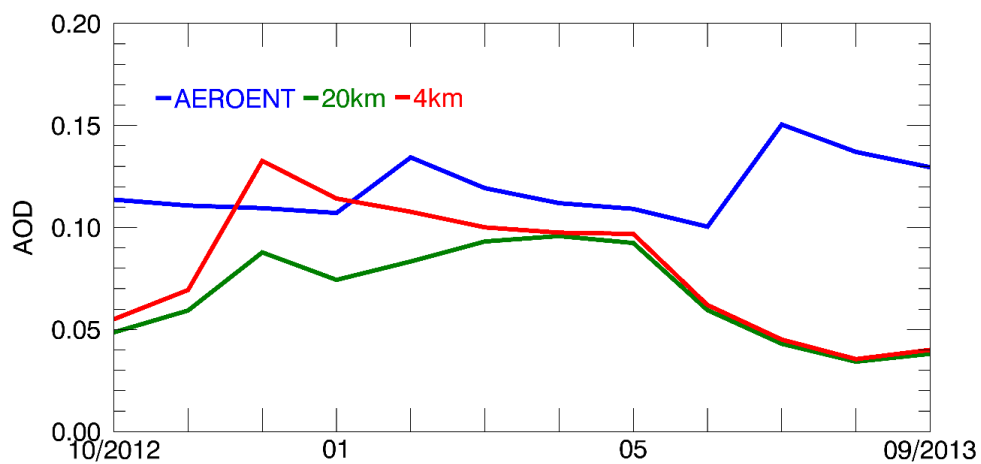
617

618 Figure 1. Daily mean anthropogenic PM_{2.5} emission rate ($\mu\text{g m}^{-2} \text{s}^{-1}$) at (a) 20km and (b) 4km
619 simulation. Red dashed lines in Figure 1a represent the region used for domain averages in Figure
620 8 and 9. Yellow circle: IMPROVE site; yellow diamond: EPA CSN site. Three urban sites: Fresno,
621 Bakersfield and Modesto; two rural sites: Pinnacles and Kaiser.



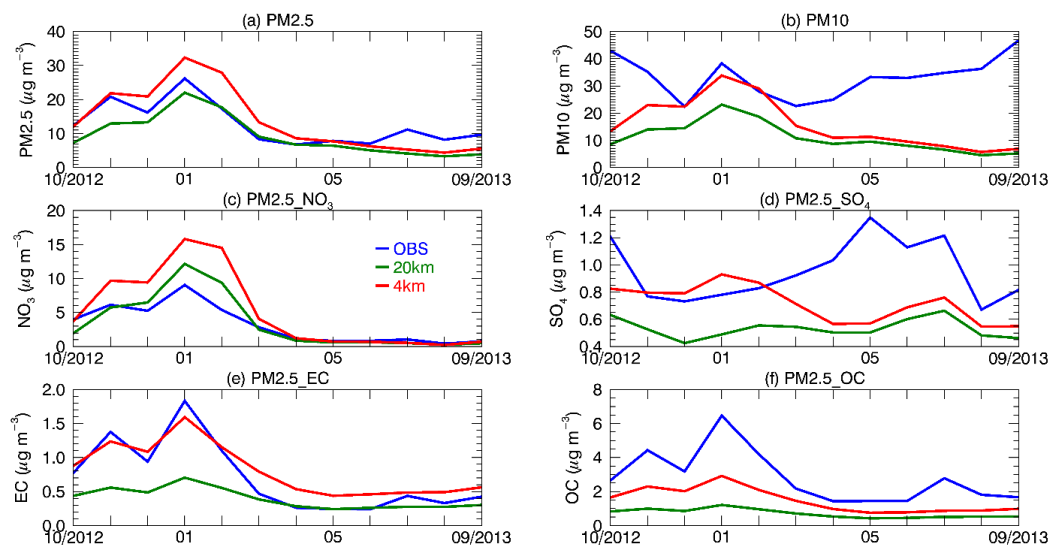
622

623 Figure 2. Spatial distribution of seasonal mean 550 nm AOD from MISR and the WRF-Chem
624 (20km and 4km) simulations in WY2013. OND: October, November and December; JFM: January,
625 February and March; AMJ: April, May and June; JAS: July, August and September.



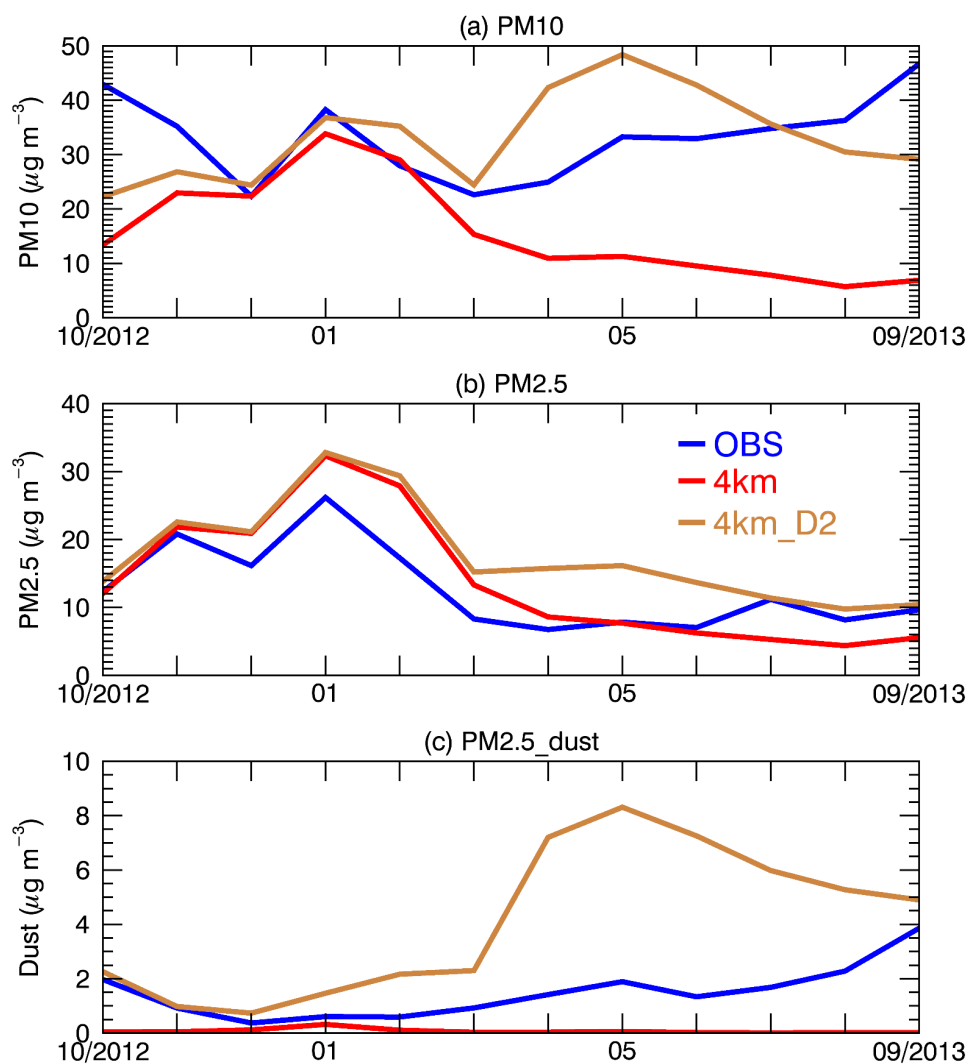
626

627 Figure 3. Monthly mean 550 nm AOD at Fresno, CA from October 2012 to September 2013.



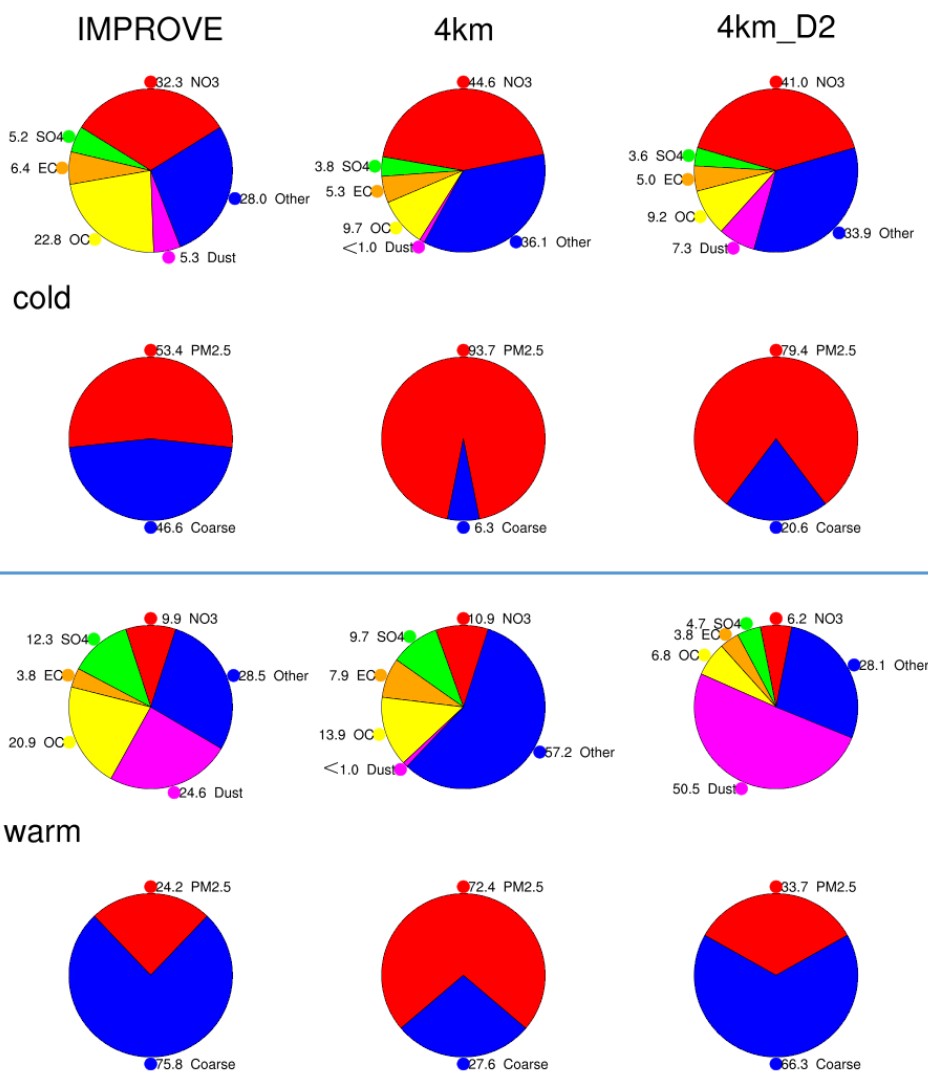
628

629 Figure 4. Aerosol mass ($\mu\text{g m}^{-3}$) for different species from IMPROVE (OBS), 20km
630 simulations at Fresno, CA. PM2.5_NO₃ represents NO₃ with diameter $\leq 2.5 \mu\text{m}$. Similar definition
631 for SO₄, EC and OC in the figures.



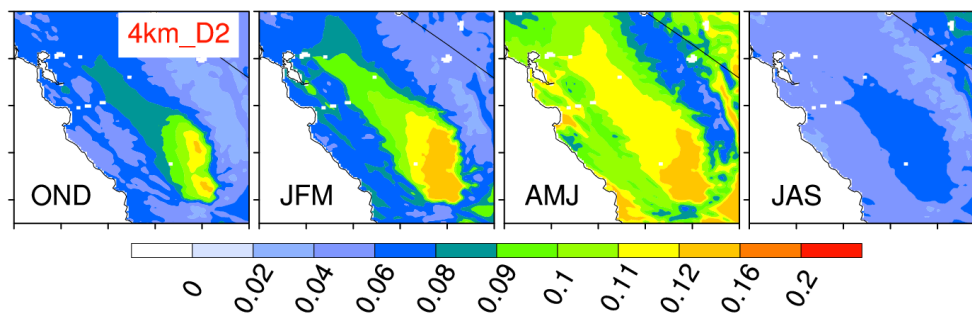
632

633 Figure 5. (a) PM10; (b) PM2.5; (c) PM2.5_dust from IMPROVE (OBS), 4km and 4km_D2
634 simulations at Fresno, CA.



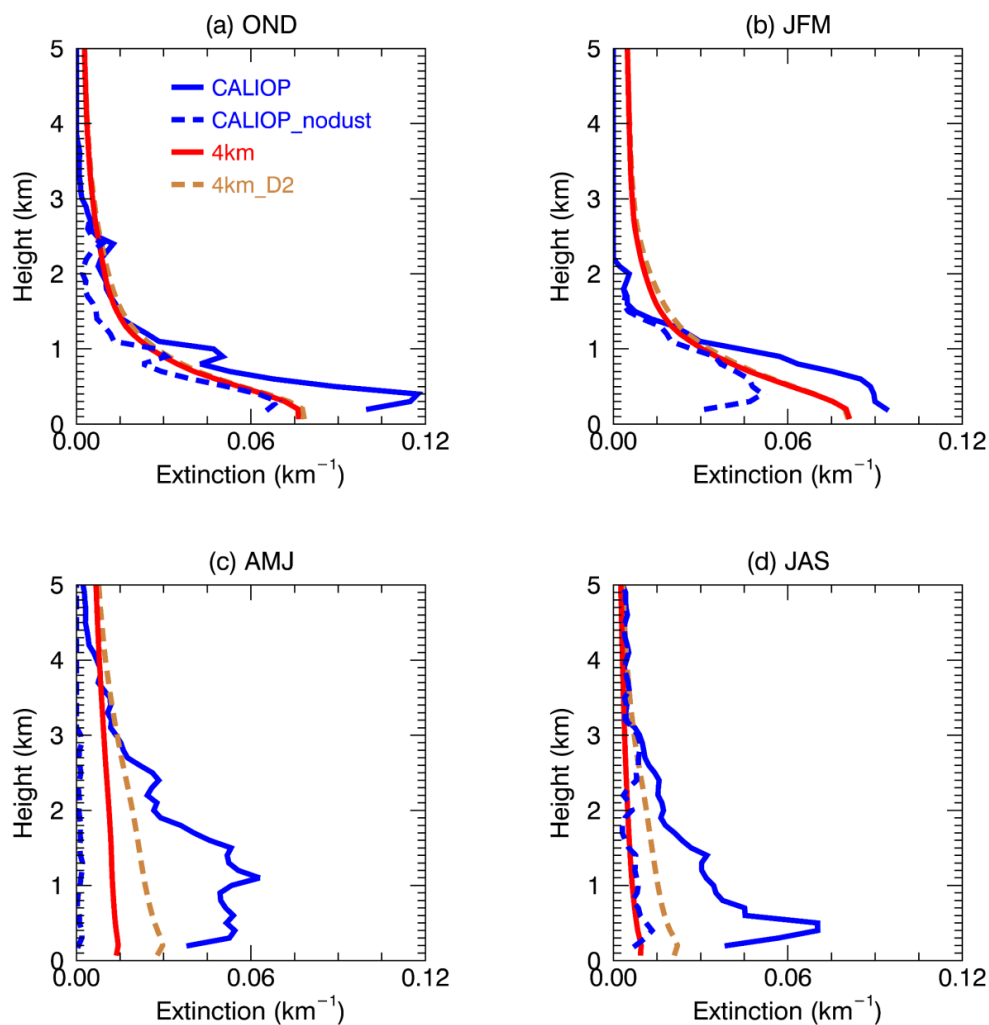
635

636 Figure 6. Relative contribution (%) of aerosol species from IMPROVE and the WRF-Chem
 637 simulations (4km and 4km_D2) at Fresno, CA in WY2013. (Panel 1) Contribution to PM2.5 in
 638 cold season; (Panel 2) relative contribution of PM2.5 and coarse mass to PM10 in cold season;
 639 (Panel 3) same as Panel 1 but in warm season; (Panel 4) same as Panel 2 but in warm season.



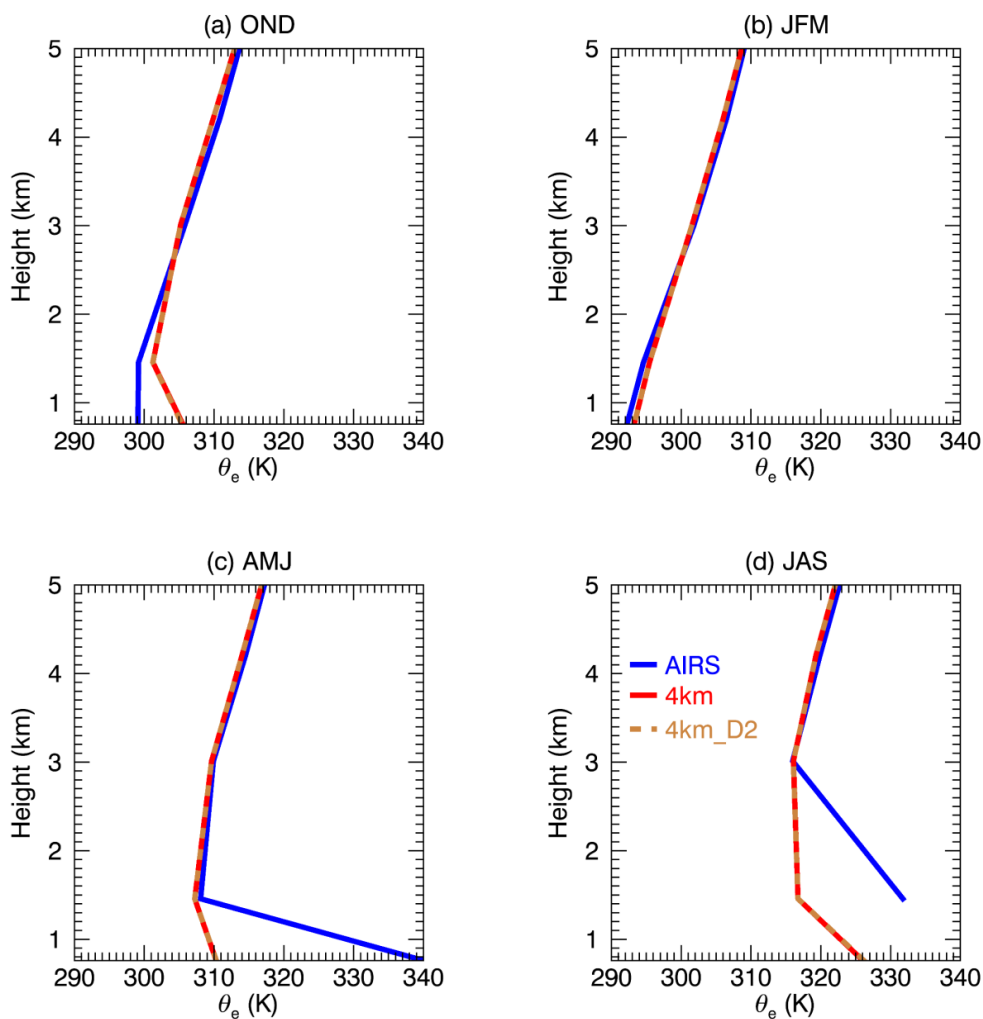
640

641 Figure 7. Spatial distribution of seasonal mean 550 nm AOD from 4km_D2 in WY2013.



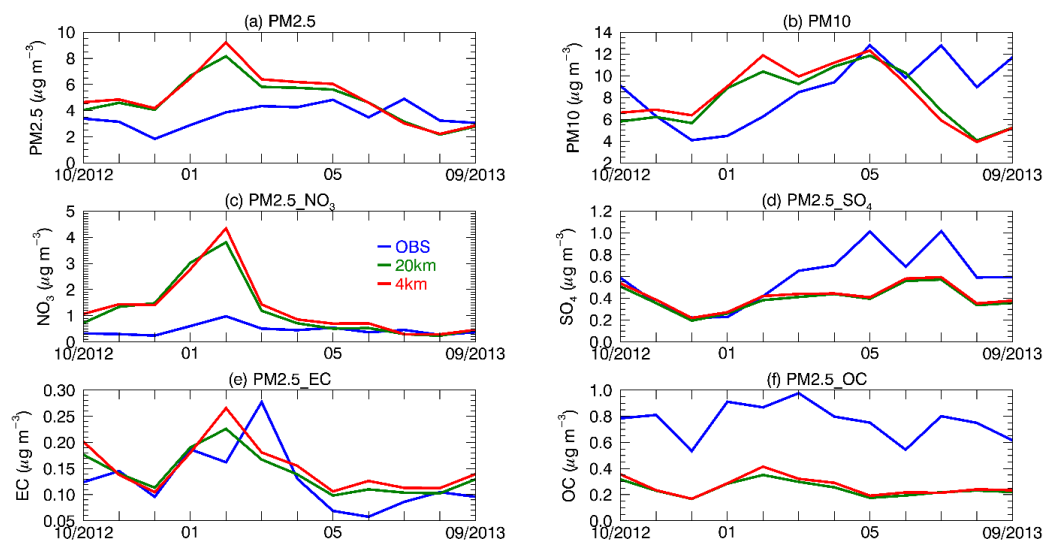
642

643 Figure 8. Vertical distribution of seasonal mean 532 nm aerosol extinction coefficient (km^{-1})
644 from CALIOP (blue) and the WRF-Chem (4km and 4km_D2) simulations over the red box
645 region in Figure 1a) in WY2013. Blue dashed lines (CALIOP_nodust) represent the CALIOP
646 profiles without dust (dust and polluted dust).



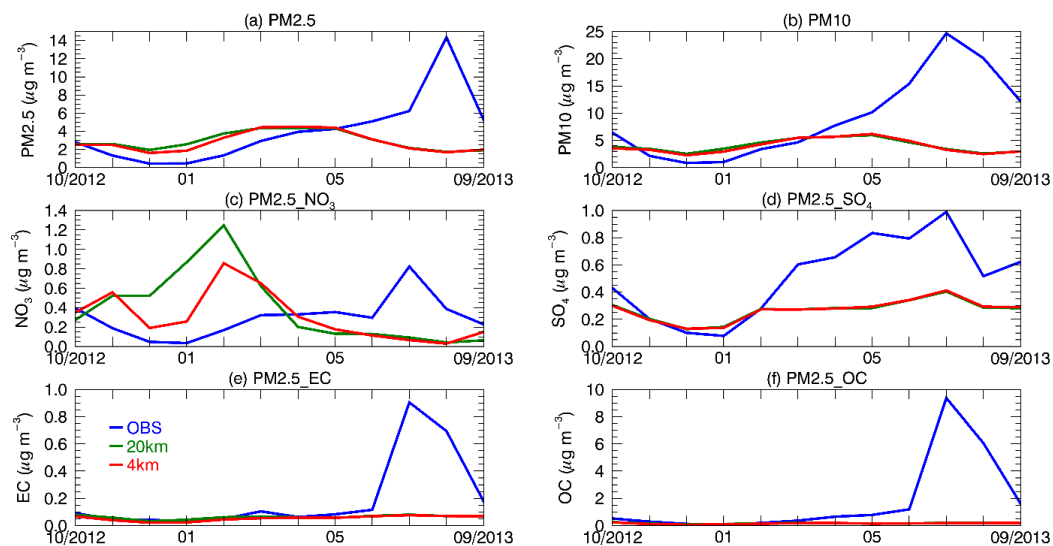
647

648 Figure 9. Vertical distribution of season mean equivalent potential temperature (θ_e ; K) from AIRS
649 and the WRF-Chem (4km and 4km_D2) simulations over the red box region in Figure 1a in
650 WY2013.



651

652 Figure 10. Aerosol mass ($\mu\text{g m}^{-3}$) for different species from IMPROVE (OBS), 20km
653 simulations at Pinnacles, CA.



654

655 Figure 11. Aerosol mass ($\mu\text{g m}^{-3}$) for different species from IMPROVE (OBS), 20km and 4km
656 simulations at Kaiser, CA.