1	The impact of aerosol optical depth assimilation on aerosol forecasts and radiative
2	effects during a wild fire event over the United States
3	
4	Dan Chen ¹ , Zhiquan Liu ¹ *, Craig S. Schwartz ¹ , Hui-Chuan Lin ¹ , Jeffrey D. Cetola ² , Yu Gu ³ and
5	Lulin Xue ¹
6	1. National Center for Atmospheric Research, Boulder, Colorado, USA
7	2. Air Force Weather Agency, Omaha, Nebraska, USA
8	3. University of California, Los Angeles, Los Angeles, California, USA
9	
10	
11	
12	Submitted to Geoscientific Model Development
13 14	Revised in September 2014
15	
16	
17	
18	
19	
20	
21	
22	

*Corresponding author: Dr. Zhiquan Liu (liuz@ucar.edu)

24 Abstract

25 The Gridpoint Statistical Interpolation three-dimensional variational data assimilation (DA) 26 system coupled with the Weather Research and Forecasting/Chemistry (WRF/Chem) model was 27 utilized to improve aerosol forecasts and study aerosol direct and semi-direct radiative feedbacks 28 during a U.S. wild fire event. Assimilation of MODIS total 550 nm aerosol optical depth (AOD) 29 retrievals clearly improved WRF/Chem forecasts of surface PM_{2.5} and organic carbon (OC) compared to the corresponding forecasts without aerosol data assimilation. The scattering 30 31 aerosols in the fire downwind region typically cooled layers both above and below the aerosol 32 layer and suppressed convection and clouds, which led to an average 2% precipitation decease 33 during the fire week. This study demonstrated that even with no input of fire emissions, AOD 34 DA improved the aerosol forecasts and allowed a more realistic model simulation of aerosol 35 radiative effects.

38 1. Introduction

Aerosols are known to affect weather and climate by modulating radiation in the atmosphere by either scattering or absorption of sunlight (direct effect, e.g. *Rosenfeld et al.*, 2008); thermodynamic effect on clouds (semi-direct, e.g. *Hansen et al.*, 1997); and altering cloud microphysical processes (indirect effects, e.g. *Kaufman and Koren*, 2006). Aerosols can scatter incoming solar radiation and cool both the surface and atmosphere (*Charlson et al.*, 1992; Kiehl and *Briegleb*, 1993). Conversely, absorbing aerosols, such as black carbon (BC) and dust can absorb solar radiation, which heats the local atmosphere (*Hansen et al.*, 1997).

One of the most important short-term effects of aerosols is the impact on local 46 47 meteorological conditions, especially clouds and precipitation. These changes can be particularly 48 pronounced during biomass burning events when large amount of aerosols are injected into the 49 atmosphere (e.g. Koren et al. 2004; Wilcox et al., 2012). Several observational studies have 50 shown evidence for aerosol-induced intensification and weakening of convection with a critical 51 aerosol optical depth (AOD) value (~0.2-0.4), below which additional aerosol enhances 52 convection and precipitation but above which additional aerosol weakens convection and 53 precipitation (Koren et al., 2008; Rosenfeld et al., 2008). For example, Koren et al. (2004) 54 analyzed Moderate Resolution Imaging Spectroradiometer (MODIS) satellite data over the 55 Amazon region during the biomass burning season and found that smoke reduced cumulus cloud 56 cover from 38% in clean conditions (AOD of ~ 0.1) to 0% in heavy smoke (AOD of ~ 1.3). 57 Andreae et al. (2004) used in situ measurements of cloud condensation nuclei and cloud droplets 58 over the Amazon and found that the suppression of low-level rainout by biomass burning smoke 59 tended to invigorate deep convective clouds, thus increasing precipitation. In addition, aerosol-60 induced changes in the atmosphere may exert different effects on clouds depending on the type

of aerosols (absorbing or scattering) and the vertical distributions of aerosols and clouds (e.g. *Rosenfeld et al.*, 2008).

63 To accurately simulate aerosol effects, it is necessary to precisely simulate aerosol types and 64 distributions. AOD Data assimilation (DA), combining satellite derived AOD observations with 65 numerical model output, has proved to be skillful at improving aerosol and AOD forecasts (e.g., 66 Collins et al., 2001; Liu et al., 2011). Liu et al. (2011, hereafter L11) implemented AOD DA 67 within the National Centers for Environmental Prediction (NCEP) Gridpoint Statistical 68 Interpolation (GSI) three-dimensional variational (3DVAR) DA system coupled to the Goddard 69 Chemistry Aerosol Radiation and Transport (GOCART) (Chin et al., 2000 and 2002) aerosol 70 scheme within the Weather Research and Forecasting/Chemistry (WRF/Chem) model (Grell et 71 al., 2005). Verification results demonstrated improved aerosol forecasts from AOD DA over a 72 week-long period while studying a dust storm in East Asia. This aerosol DA system was also 73 used to assimilate surface PM_{2.5} over the U. S. (Schwartz et al., 2012, hereafter S12) and PM₁₀ 74 over China (Jiang et al., 2013).

75 These previous air-quality oriented studies (L11; S12; Jiang et al., 2013) illustrated the ability 76 of aerosol DA to improve forecasts of total aerosol mass in terms of AOD, PM_{2.5} and PM₁₀, but 77 did not verify aerosol speciation forecasts. As pointed out in L11, the aerosol data assimilation 78 system used here directly analyzes 3D mass concentration of individual aerosol species and 79 allows them to adjust independently with additional constraint from the background error 80 covariance for individual species. Similar method was also adopted by Kahnert (2009) for 81 aerosol inverse modeling. This work builds upon L11 and S12 and serves two purposes. First, 82 this study aims to verify the GSI 3DVAR DA system's capability to analyze and forecast aerosol 83 species, including black carbon (BC) and organic carbon (OC), during a fire event without fire emission input in the WRF/Chem model. Second, the biomass burning aerosol radiative effects
(direct and semi-direct) on clouds and precipitation in the downwind region during the fire event
are investigated.

87 2. Model Description and Experimental Design

88 Version 3.4.1 of WRF/Chem was used and configurations mostly followed S12. The model 89 domain (operationally used at the US Air Force Weather Agency) with 20-km horizontal grid 90 spacing covered a large portion (20-degree north) of the Northern Hemisphere with the polar 91 projection (not shown), although our analysis will focus on North American regions where a 92 wild fire occurred (Fig. 1). There were 57 vertical levels extending from the surface to 10 hPa. 93 Aerosol direct and semi-direct effects were implemented (Fast et al., 2006) in WRF/Chem by 94 linking the optical properties of simulated GOCART aerosols (OC, BC, sulfate, dust and sea salt) to the Goddard Space Flight Center Shortwave radiation scheme (Chou and Suarez, 1994). 95 96 Aerosol optical properties, including scattering/absorption coefficients and single-scattering 97 albedos, are calculated by the "aerosol chemical to aerosol optical properties" module built in 98 WRF/Chem (Fast et al., 2006; Barnard et al., 2010). Aerosol indirect effects were not 99 implemented for GOCART with the WRF/Chem version used. The WRF single-moment 6-class 100 microphysics scheme and the Grell-Devenyi ensemble cumulus scheme (Grell and Devenyi, 101 2002) were used. Anthropogenic emissions were provided by the 0.5×0.5 degree Reanalysis of 102 the TROpospheric (RETRO) chemical composition the 40 over past vears 103 (http://retro.enes.org/data emissions.shtml) and the 0.1×0.1 degree Emission Database for 104 Global Atmospheric Research (EDGAR) (http://themasites.pbl.nl/tridion/en/themasites/edgar/). 105 Over the U.S., the high resolution (4-km) National Emission Inventory 2005 (NEI'05) emission 106 was used for more accuracy (Kim et al., 2011). Within WRF/Chem, Emissions of dust and seasalt were parameterized using the GOCART dust and sea-salt modules (*Chin et al.*, 2002). The
lateral boundary conditions (LBCs) for meteorological fields were provided by the NCEP Global
Forecast System (GFS). LBCs for chemistry/aerosol fields were idealized profiles embedded
within the WRF/Chem model as in S12.

111 To evaluate the GSI-WRF/Chem system's capability of improving aerosol species and 112 simulating aerosol radiative effects during the fire event, which originated in the western U.S. 113 and smoke Aug. 2012 sent eastward during 13-18. 114 (http://earthobservatory.nasa.gov/IOTD/view.php?id=78881&src=ve), two DA experiments 115 were conducted. One experiment assimilated only NCEP conventional meteorological 116 observations (MET) while the other assimilated both meteorological data and MODIS level-2 117 (10km×10km resolution) 550 AOD retrievals obtained from nm 118 ftp://ladsweb.nascom.nasa.gov/allData/51/MOD06 L2 (and MYD06 L2) (MET AOD). Only 119 the AOD data flagged as the best quality were used in this study. Each experiment started 120 WRF/Chem simulation with a 6-h cycling interval from 00 UTC 1 August in order to spin up 121 aerosol fields before the fire event. For MET, GSI 3DVAR meteorological (surface pressure, 3D 122 wind, temperature and moisture) analyses (Wu et al., 2012) were performed using the previous 123 cycle's 6 h forecast (meteorological fields only) as the background, and aerosol fields were 124 simply carried over from cycle to cycle (similar to a continuous aerosol forecast). For 125 MET AOD, GSI 3DVAR updated both meteorological and GOCART aerosol variables (only at 126 18 UTC when AOD data were available over US) every 6 h, again using the previous cycle's 6 h 127 forecast as the background. The assimilation time window was ± 1.5 -h centered at analysis times 128 (00, 06, 12, and 18 UTC). This cyclic experimental design was also adopted by L11 and S12, 129 who assimilated aerosol observations only. No cross-correlation between meteorological and

aerosol fields was allowed in MET_AOD even though meteorological and AOD data wereassimilated simultaneously. More details related to AOD DA can be found in L11 and S12.

This design permitted a clear isolation of the impact of AOD DA. To investigate aerosol radiative effects, 48 h forecasts were initialized at 00 UTC for each experiment during the fire week. Hourly model outputs were analyzed. Since the meteorological fields after 3DVAR DA in the two experiments were very close, the forecast differences of meteorological fields suggest primarily radiative effects due to fire emitted aerosols.

137 3. PM Speciation Verification

Surface observations, including hourly $PM_{2.5}$ from the EPA AIRNow network and 24haveraged BC and OC (available every three days) from the Interagency Monitoring of PROtected Visual Environments (IMPROVE) network, were used for aerosol verification. Figure 1 shows the locations of these sites. The averaged AOD differences between the two experiments (MET_AOD minus MET) for the fire period (Aug 14-17) are also shown in Fig 1. Significant increases in AOD (~0.4) over the western U.S. and the fire downwind region (FDR, indicated by the red rectangle in Fig. 1) were produced after assimilating MODIS AOD.

145 Figure 2 shows the average PM_{2.5}, BC and OC observations and model forecasts between 146 August 1-22, 2012 over the sites located in the fire originating area (western U.S. 130-105°W) 147 and fire downwind regions (eastern U.S. 105-70°W). Model outputs from the two experiments were interpolated to the observation sites. The 6-h WRF/Chem forecasts of $PM_{2.5}$ were compared 148 149 with AIRNow observations at 00, 06, 12, 18 UTC. To compare the forecasts with IMPROVE 150 24h-averaged (from 06 to 06 UTC) BC and OC observations, the corresponding 6-h model 151 forecasts were also averaged. Observations (black lines) show large peaks in total PM2.5, BC and 152 OC during the fire event (Aug. 13-16) in the western U.S., due to strong fire emissions. While the experiment without AOD DA (blue lines) failed to reproduce those peaks and underpredicted aerosol concentrations, most likely a result of the lack of fire emission input in the model, the experiment with AOD DA (red lines) substantially improved surface PM_{2.5} forecasts. Furthermore, the peaks of individual aerosol species' concentrations (especially OC) were well captured with AOD DA, although OC and BC were still underpredicted when the maximum concentrations were reached on 13 August in the Western U.S.

159 Observations also show increased total PM_{2.5} and OC in the downwind region when the smoke 160 was transported eastward during the fire event. MET AOD improved substantially the 161 simulation with increased OC and PM_{2.5} when compared with MET. While MET exhibits a 162 relatively small bias for BC, large low biases can be seen for PM_{2.5} and OC in both regions even 163 during periods without fire, which may indicate model deficiencies related to emissions and 164 other physical/chemical processes. AOD DA helped correct these biases and improved the 165 simulation for the total mass (i.e., PM_{2.5}) and for OC (and to a lesser extent for BC in the 166 Western U.S.) in this case.

167 4. Aerosol Radiative Feedback

168 Fire emitted aerosols scatter and absorb solar radiation in daytime and thus can affect the 169 atmospheric temperature profiles. Averaged over the FDR region, which was cloudier than the 170 Western U.S. during the fire period and thus cloud/precipitation features were more likely to be 171 modified through aerosol semi-direct effects, the time series of hourly model outputs of day-2 172 forecasts (i.e., 24h-47h forecasts valid from 00 to 23 UTC) of 550-nm AOD and shortwave 173 downward fluxes reaching the surface (SWDOWN) from the two experiments are shown in Fig 174 3a-b. The jumps in AOD values from 23 UTC to 00 UTC are most likely the result of forecast 175 range differences (i.e., 47h vs. 24h forecast). The average AOD differences reach as high as

0.16-0.20 on Aug. 17, which is almost 80% of the total AOD from the MET_AOD experiment.
The average AOD differences were around 0.08 after Aug. 20 when fire emissions decreased.
The AOD increase led to more aerosol scattering and absorption in MET_AOD, which resulted
in a SWDOWN reduction of ~10 w/m² during Aug. 15-18 with much smaller changes afterward.
Also note that small SWDOWN differences occurred in the late afternoon of Aug. 15, which was
likely caused by cloud feedback.

182 Similar to Fig. 3, Fig. 4 shows the FDR-averaged differences of 550-nm AOD, temperature, 183 relative humidity, vertical velocity, cloud liquid and cloud ice water as a function of height and 184 time (hourly output of day-2 forecasts) between the two experiments. The largest AOD (also OC 185 and BC, not shown) increase due to AOD DA occurred at around 4-5 km altitude, indicating 186 upward transport of fire emitted aerosols. This peak AOD height in the AOD DA experiment is 187 consistent with the altitude at which OC and BC had maximum background error variances (not 188 shown). The decreased temperature below this level indicates that the additional aerosols cooled 189 the surface layer and planetary boundary layer (PBL, ~2 km in the afternoon). A weak cooling 190 appeared above the aerosol layer and a weak warming was noted around 15 km. Temperature 191 changed little in the aerosol layer, as the absorbing aerosols (BC and dust) were not dominant in 192 the FDR and no obvious differences of those species were evident between the two experiments 193 (not shown). The relative humidity differences roughly followed the temperature differences, 194 with increased RH in the PBL and above the aerosol layer. Cooler and moister air in the PBL 195 (below ~2km) facilitates low cloud formation from MET AOD simulations (Aug 17-19), which 196 was especially pronounced on Aug. 17 when the AOD increase reached its maximum. Middle 197 level liquid clouds above the PBL and below the aerosol layer decreased, likely associated with 198 deceased relative humidity. The ice clouds near the tropopause also decreased, which may be related to the suppression of upward motion in the middle and upper troposphere (Fig. 4b). The aerosol direct and semi-direct effects are consistent with *Jacobson* (2005) and the findings of middle and high cloud suppression are similar to Amazon fire events (*Koren et al.* 2004; *Wu et al.*, 2012).

203 Figure 3c shows the average precipitation differences (red line, left Y-axis) between the two 204 experiments in the FDR and the corresponding total amount of precipitation (mm/grid) from 205 model forecasts and Stage IV observations (black lines, right Y-axis). Surface precipitation was 206 suppressed: precipitation decreased by up to 0.03 mm/grid (7.3%) late on 16 August and the 207 average precipitation during the fire week was reduced by 2.0%, perhaps associated with the 208 suppressed middle clouds and ice-clouds (Fig. 4d) (Rosenfeld et al., 2008). The radiative impact 209 of aerosols on precipitation reported here is consistent with Zhao et al. (2011) and Wu et al., 210 (2012), who focused on Asian dust and Amazon fires, respectively. Overall, WRF/Chem 211 produced reasonably good precipitation forecasts when compared to Stage IV observations even 212 though the total amount was usually overpredicted.

213 **5.** Summary

The GSI 3DVAR DA system coupled with the WRF/Chem model successfully simulated surface BC, OC, and $PM_{2.5}$ during a wild fire event without any fire emission input in the model. By assimilating total 550-nm AOD retrievals from MODIS sensors, surface $PM_{2.5}$ and OC in the fire originating regions were substantially improved compared to those when AOD was not assimilated. The increased aerosols in the downwind regions were dominated by OC and other oxidized $PM_{2.5}$ components, which are mainly scattering aerosols.

Direct and semi-direct aerosol radiative effects due to aerosols in the downwind region wereinvestigated. Enhanced scattering aerosol concentrations due to AOD DA cooled layers both

below and above the aerosol layer, leading to changes in the temperature, relative humidity, vertical velocity and clouds. We found that the radiative effect of the increased AOD (varied from \sim 0.2- \sim 0.4) was to increase cloud amount in the PBL and suppress middle level liquid clouds and high level ice clouds. A 2% average reduction of total precipitation due to aerosol increase was also evident. This study demonstrated the value of aerosol DA for more accurately depicting the aerosol spatial distribution and speciation and thus allowed a more realistic model simulation of aerosol radiative effects during a fire event even with no input of fire emissions.

229 Grell et al. (2011) showed that the inclusion of fire emissions and a plume rise scheme 230 resulted in strong modifications of cloud and precipitation features in high-resolution (10km/2km 231 nested domains) WRF/Chem simulations with both direct and indirect aerosol feedbacks for a 232 wildfire event over Alaska. However, in our initial trials, the inclusion of GOES WF ABBA 233 (Geostationary Operational Environmental Satellite - Wildfire Automated Biomass Burning 234 Algorithm) (Prins et al., 1998) fire emissions in the simulation of this fire event over California 235 led to a substantial overestimation of aerosol concentrations when compared to surface PM_{25} 236 OC and BC measurements (not shown). The impact of AOD DA together with the inclusion of 237 fire emissions will be further investigated in the future.

238 239

240 Acknowledgements

- 241
- 242 This work is supported by grants from the U.S. Air Force Weather Agency. NCAR is sponsored
- 243 by the National Science Foundation.

244

```
245 References
```

246

249 10.1126/science.1092779, 2004.

²⁴⁷ Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M., and Silva-

²⁴⁸ Dias, M. A. F.: Smoking rain clouds over the Amazon, Science, 303, 1337-1342, DOI

- 250
- Barnard, J. C., Fast, J. D., Paredes-Miranda, G., Arnott, W. P., and Laskin, A.: Technical Note:
 Evaluation of the WRF-Chem "Aerosol Chemical to Aerosol Optical Properties" Module using
 data from the MILAGRO campaign, Atmos Chem Phys, 10, 7325-7340, DOI 10.5194/acp-107325-2010, 2010.
- 255
- Charlson, R. J., Schwartz, S. E., Hales, J. M., Cess, R. D., Coakley, J. A., Hansen, J. E., and
 Hofmann, D. J.: Climate Forcing by Anthropogenic Aerosols, Science, 255, 423-430, DOI
 10.1126/science.255.5043.423, 1992.
- 259

Chin, M., Savoie, D. L., Huebert, B. J., Bandy, A. R., Thornton, D. C., Bates, T. S., Quinn, P. K.,
Saltzman, E. S., and De Bruyn, W. J.: Atmospheric sulfur cycle simulated in the global model
GOCART: Comparison with field observations and regional budgets, J Geophys Res-Atmos,
105, 24689-24712, Doi 10.1029/2000jd900385, 2000.

- 264
- 265 Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B. N., Duncan, B. N., Martin, R. V., Logan,
- 266 J. A., Higurashi, A., and Nakajima, T.: Tropospheric aerosol optical thickness from the
- GOCART model and comparisons with satellite and Sun photometer measurements, J Atmos
 Sci, 59, 461-483, Doi 10.1175/1520-0469(2002)059<0461:Taotft>2.0.Co;2, 2002.
- 269
- Chou, M.-D., and M.J. Suarez, An efficient thermal infrared radiation parameterization for use in
 general circulation models, NASA Tech. Memo., TM 104606, vol. 3, 25 pp., NASA Goddard
 Space Flight Cent., Greenbelt, Md, 1994.
- 273
- Collins, W. D., Rasch, P. J., Eaton, B. E., Khattatov, B. V., Lamarque, J. F., and Zender, C. S.:
 Simulating aerosols using a chemical transport model with assimilation of satellite aerosol
 retrievals: Methodology for INDOEX, J Geophys Res-Atmos, 106, 7313-7336, Doi
 10.1029/2000jd900507, 2001.
- 278

Fast, J.D., Gustafson, W.I., Easter, R.C., Zaveri, R.A., Barnard, J.C., Chapman, E.G.,
Grell, G.A., Peckham, S.E.: Evolution of ozone, particulates, and aerosol direct radiative
forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol
model, J. Geophys Res-Atmos, 111, D21305. Doi 10.1029/2005jd006721. 2006.

- 283 284
- Grell, G. A., and Devenyi, D.: A generalized approach to parameterizing convection combining
 ensemble and data assimilation techniques, Geophys Res Lett, 29, Artn 1693, Doi
 10.1029/2002gl015311, 2002.
- 288
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and
 Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmos Environ, 39, 69576975, DOI 10.1016/j.atmosenv.2005.04.027, 2005.
- 292
- 293 Grell, G., Freitas, S. R., Stuefer, M., and Fast, J.: Inclusion of biomass burning in WRF-Chem:
- impact of wildfires on weather forecasts, Atmos Chem Phys, 11, 5289-5303, DOI 10.5194/acp11-5289-2011, 2011.

- 296
- Hansen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response, J Geophys Res-
- Atmos, 102, 6831-6864, Doi 10.1029/96jd03436, 1997.
- Jacobson, M. Z.: Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming (vol 107, pg 4410, 2002), J Geophys Res-
- 302 Atmos, 110, Artn D14105, Doi 10.1029/2004jd005888, 2005.
- 303
- Jiang, Z. Q., Liu, Z. Q., Wang, T. J., Schwartz, C. S., Lin, H. C., and Jiang, F.: Probing into the
- 305 impact of 3DVAR assimilation of surface PM10 observations over China using process analysis,
- 306 J Geophys Res-Atmos, 118, 6738-6749, Doi 10.1002/Jgrd.50495, 2013.
- Kahnert, M.: On the observability of chemical and physical aerosol properties by optical
 observations: Inverse modeling with variational data assimilation. Tellus 61B, 747-755, 2009
- Kaufman, Y. J., and Koren, I.: Smoke and pollution aerosol effect on cloud cover, Science, 313,
 655-658, DOI 10.1126/science.1126232, 2006.
- 311
- Kiehl, J. T., and Briegleb, B. P.: The Relative Roles of Sulfate Aerosols and Greenhouse Gases
 in Climate Forcing, Science, 260, 311-314, DOI 10.1126/science.260.5106.311, 1993.
- 314
- 315 Kim, S. W., McKeen, S. A., Frost, G. J., Lee, S. H., Trainer, M., Richter, A., Angevine, W. M.,
- Atlas, E., Bianco, L., Boersma, K. F., Brioude, J., Burrows, J. P., de Gouw, J., Fried, A.,
- 317 Gleason, J., Hilboll, A., Mellqvist, J., Peischl, J., Richter, D., Rivera, C., Ryerson, T., Hekkert, S.
- T. L., Walega, J., Warneke, C., Weibring, P., and Williams, E.: Evaluations of NOx and highly
- 319 reactive VOC emission inventories in Texas and their implications for ozone plume simulations
- during the Texas Air Quality Study 2006, Atmos Chem Phys, 11, 11361-11386, DOI
- 321 10.5194/acp-11-11361-2011, 2011.
- 322
- 323 Koren, I., Kaufman, Y. J., Remer, L. A., and Martins, J. V.: Measurement of the effect of
- Amazon smoke on inhibition of cloud formation, Science, 303, 1342-1345, DOI
- 325 10.1126/science.1089424, 2004.
- 326
- Koren, I., Martins, J. V., Remer, L. A., and Afargan, H.: Smoke invigoration versus inhibition of
 clouds over the Amazon, Science, 321, 946-949, DOI 10.1126/science.1159185, 2008.
- Liu, Z. Q., Liu, Q. H., Lin, H. C., Schwartz, C. S., Lee, Y. H., and Wang, T. J.: Three-
- dimensional variational assimilation of MODIS aerosol optical depth: Implementation and
- application to a dust storm over East Asia, J Geophys Res-Atmos, 116, Artn D23206
- 333 Doi 10.1029/2011jd016159, 2011.
- 334
- Prins, E. M., Feltz, J. M., Menzel, W. P., and Ward, D. E.: An overview of GOES-8 diurnal fire
 and smoke results for SCAR-B and 1995 fire season in South America, J Geophys Res-Atmos,
 103, 31821-31835, Doi 10.1029/98id01720, 1998.
- 338

- 339 Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A.,
- and Andreae, M. O.: Flood or drought: How do aerosols affect precipitation?, Science, 321,
- 341 1309-1313, DOI 10.1126/science.1160606, 2008.
- 342
- 343 Schwartz, C. S., Liu, Z. Q., Lin, H. C., and McKeen, S. A.: Simultaneous three-dimensional
- variational assimilation of surface fine particulate matter and MODIS aerosol optical depth, J
 Geophys Res-Atmos, 117, Artn D13202
- 346 Doi 10.1029/2011jd017383, 2012.
- 347
- Wilcox, E. M.: Direct and semi-direct radiative forcing of smoke aerosols over clouds, Atmos
 Chem Phys, 12, 139-149, DOI 10.5194/acp-12-139-2012, 2012.
- 350
- Wu, W. S., Purser, R. J., and Parrish, D. F.: Three-dimensional variational analysis with spatially
 inhomogeneous covariances, Mon Weather Rev, 130, 2905-2916, Doi 10.1175/15200493(2002)130<2905:Tdvaws>2.0.Co;2, 2002.
- 354
- 355 Wu, L. T., Su, H., and Jiang, J. H.: Regional simulations of deep convection and biomass
- burning over South America: 2. Biomass burning aerosol effects on clouds and precipitation, J
- 357 Geophys Res-Atmos, 116, Artn D17209
- 358 Doi 10.1029/2011jd016106, 2011.359
- Zhao, C., Liu, X., Leung, L. R., and Hagos, S.: Radiative impact of mineral dust on monsoon
 precipitation variability over West Africa, Atmos Chem Phys, 11, 1879-1893, DOI 10.5194/acp11-1879-2011, 2011.
- 363
- 364
- 365
- 366
- 367
- 368
- 369
- 370
- 370
- 371
- 372



Figure 1. The domain for aerosol verification. The mean AOD difference between the two
experiments (MET_AOD minus MET, see text in section 2) for Aug. 14-17, 2012. The locations
of AIRNow (open circle) and IMPROVE (dot) sites are also shown. The red rectangle is defined
as the fire downwind region (FDR) used in the radiative effect analysis.



Figure 2. The time series of model predicted (6-h forecasts) and observed PM_{2.5}, BC and OC,
averaged over the (a) western (130-105°W) and (b) eastern U.S. (105-70°W) during Aug. 2012.
PM_{2.5} is in 6-h interval. BC and OC are in 72-h interval.



Figure 3. The hourly model output of day-2 forecasts averaged over the FDR for (a) 550-nm AOD, (b) shortwave downward fluxes and (c) precipitation during Aug. 15-21. Red lines: the difference of MET_AOD minus MET (left *Y*-axis); Black lines: the total amount from MET_AOD (right *Y*-axis).



Figure 4. Similar to Fig. 3, but for the FDR averaged differences of MET_AOD minus MET for
(a) AOD, (b) vertical velocity, (c) temperature (contours) and relaive humidity (color shaded)
and (d) liquid and ice clouds as a function of height and time.