Vertical Accumulation of Ozone and Aerosol during the 2016 Southeastern U.S. Wildfires

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Abstract

The vertical accumulation of ozone and aerosol during an episode of the 2016 Southeastern United States Wildfires is analyzed by integrating a regional chemical transport model with ozonesonde, O\$.3\$ Differential Absorption Lidar (DIAL), ceilometer, surface monitors, and satellite products. The results indicate that measurements capture the vertical extent of the smoke plumes affecting the surface and upper air over Huntsville, AL, and also the enhanced ozone lamina in the plumes. Sensitivity simulations and tendency diagnostics characterize the chemical and physical processes affecting the vertical profiles downstream of the wildfires. The model results show that the net chemical ozone production (PO\$.3\$) dominates the daytime ozone accumulation by up to 19 ppb/10 hrs in the upper air over Huntsville. At the surface, the negative PO\$.3\$ is offset by positive O\$.3\$ contributions from vertical mixing and advection. Fire emissions increase the vertical ozone by affecting local chemical reactions, transportation, and vertical exchange. The dominant processes exhibit daily, diurnal, and vertical variability. Quantitatively, fire emissions increase the daytime positive PO\$.3\$ by up to 25\% in the upper air, and increase the daytime PM2.5 by up to 77\%. The capability of the regional model for reproducing the observations is explored. Increasing the fire aerosol emissions improves the model performance on domain-averaged PM2.5. The model captures the well-mixed aerosol in the boundary layer but fails to fully reproduce the densest plumes seen in the DIAL and satellite. The discrepancies are associated with poor satellite observing condition due to clouds and with uncertainties in emission inventories.

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Key Points: 10

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11	•	WRF-Chem performance and emission inputs are evaluated against ozonesonde,
12		UV DIAL, EPA PM2.5, EPA O_3 , MODIS AOD, and fire inventories.
13	•	(OC+BC)/CO ratios in fire inventories differ by a factor of 5.7 within fire region.
14		Aerosol adjustment affects modeled PM2.5, aerosol extinction, and AOD.
15	•	Fire emissions increase daytime net chemical ozone production by up to 25% in
16		the upper air, and increase PM2.5 by up to 77% during daytime.

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17 Abstract

The vertical accumulation of ozone and aerosol during an episode of the 2016 South-18 eastern United States Wildfires is analyzed by integrating a regional chemical transport 19 model with ozonesonde, O_3 Differential Absorption Lidar (DIAL), ceilometer, surface 20 monitors, and satellite products. The results indicate that measurements capture the ver-21 tical extent of the smoke plumes affecting the surface and upper air over Huntsville, AL, 22 and also the enhanced ozone lamina in the plumes. Sensitivity simulations and tendency 23 diagnostics characterize the chemical and physical processes affecting the vertical pro-24 files downstream of the wildfires. The model results show that the net chemical ozone 25 production (PO_3) dominates the daytime ozone accumulation by up to 19 ppb/10 hrs 26 in the upper air over Huntsville. At the surface, the negative PO_3 is offset by positive 27 O_3 contributions from vertical mixing and advection. Fire emissions increase the ver-28 tical ozone by affecting local chemical reactions, transportation, and vertical exchange. 29 The dominant processes exhibit daily, diurnal, and vertical variability. Quantitatively, 30 fire emissions increase the daytime positive PO_3 by up to 25% in the upper air, and in-31 crease the daytime PM2.5 by up to 77%. The capability of the regional model for repro-32 ducing the observations is explored. Increasing the fire aerosol emissions improves the 33 model performance on domain-averaged PM2.5. The model captures the well-mixed aerosol 34 in the boundary layer but fails to fully reproduce the densest plumes seen in the DIAL 35 and satellite. The discrepancies are associated with poor satellite observing condition 36 due to clouds and with uncertainties in emission inventories. 37

38 1 Introduction

Biomass burning (BB) can release substantial aerosol and ozone (O_3) precursors 39 that affect climate and air quality Akagi et al. (2011); Andreae and Merlet (2001); Crutzen 40 and Andreae (1990); Crutzen, Heidt, Krasnec, Pollock, and Seiler (1979). In the past decades, 41 observation and modeling studies have indicated that BB emissions contributed to lo-42 cal and regional air-quality problems Baker et al. (2016); Hodzic et al. (2007); D. A. Jaffe 43 et al. (2013); Pfister, Wiedinmyer, and Emmons (2008); Wigder, Jaffe, and Saketa (2013), 44 as well as to downwind air-quality problems by long-range transport Colarco et al. (2004); 45 Cook et al. (2007); D. Jaffe et al. (2004); Lapina, Honrath, Owen, Val Martin, and Pfister (2006); Lindaas et al. (2017); Martin et al. (2006); McKeen et al. (2002); Morris et 47 al. (2006); Oltmans et al. (2010); Rogers, Ditto, and Gentner (2020); Sapkota et al. (2005). 48 The impacts of biomass burning on air quality vary dramatically over time and space. 49

Chemical transport models (CTMs) have been widely used for estimating fire im-50 pacts. CTMs can provide good spatio-temporal coverage, differentiate the impacts of spe-51 cific sources, and support mechanism understanding of chemical and dynamical processes Baker 52 et al. (2018). However, large uncertainties in fire-emission estimations and their treat-53 ment in models present challenges for estimating the variability of fire impacts. Uncer-54 tainties of fire-emission estimation can arise from inherent limitations of satellite detec-55 tion (e.g., polar-orbiting detection, cloud/haze burden, small fires) and inherent uncer-56 tainties of empirical approaches for emission estimations Carter et al. (2020); Justice et 57 al. (2002); Liu et al. (2020); Van Der Werf et al. (2017); Wang et al. (2018). The emis-58 sion estimation uncertainties can affect simulated smoke loading in domain-averaged scale, 59 and at local and hourly-to-daily scales in particular Cohen, Ng, Lim, and Chua (2018); 60 Liu et al. (2020); Zhang et al. (2014). Inappropriate model treatment can induce mis-61 placement of smoke plumes and O_3 vertically Baker et al. (2018); Cohen et al. (2018); 62 Fast et al. (2016, 2006); Wu et al. (2017). Therefore, observation evaluation is essential 63 to understand the model bias and emission uncertainties for a given fire event. 64

Although satellites and surface monitors make routine measurements of atmospheric O_3 concentration, balloon soundings and the lidar technique can provide precise vertically resolved O_3 observations throughout the troposphere and lower stratosphere Thompson et al. (2011). This vertical information significantly benefits air-quality management

- and modeling improvement Cooper, Langford, Parrish, and Fahey (2015). We take ad-
- vantage of both ozonesonde Newchurch, Ayoub, Oltmans, Johnson, and Schmidlin (2003)
- and ozone lidar Kuang, Burris, Newchurch, Johnson, and Long (2011) at the University
- of Alabama in Huntsville (UAH) to observe vertical profiles. The UAH ozone lidar is af-
- filiated with the Tropospheric Ozone Lidar Network (TOLNet, https://www-air.larc.nasa.gov/missions/TOLNet/).
- ⁷⁴ Under a collaborative protocol, the TOLNet lidars have demonstrated their feasibility
- and capability in fire studies M. Johnson, Kuang, Wang, and Newchurch (2016); Kuang
- et al. (2017); A. Langford et al. (2015); Reid et al. (2017); Strawbridge et al. (2018) and
- extensive scientific projects Gronoff et al. (2019); Leblanc, Brewer, Wang, and Grana-
- dos Muñoz (2018); Sullivan et al. (2019). The continuous profiling of ozone and aerosols
- ⁷⁹ provides details missed by isolated measurements and is an asset for model evaluation
- by coordinating measurements A. Langford et al. (2018); A. O. Langford et al. (2019).
 In addition, the ultraviolet (UV) backscatter (or extinction) profiles retrieved from ozone
- In addition, the ultraviolet (UV) backscatter (or extinction) profiles retrieved from ozolidar can quantify the aerosol variability at high spatio-temporal resolution and these
- lidar can quantify the aerosol variability at high spatio-temporal resolution and these
 measurements serve as a tracer for fire smoke Kuang et al. (2020); A. O. Langford et al.
- (n.d.). To our best knowledge, there has been little or no attempt to evaluate CTMs us-
- ing this range-resolved UV aerosol optical product.

Integrating vertical observations into CTMs can also improve our understanding 86 of the fire impacts on vertical profiles. Due to multiple O_3 sources in troposphere and 87 a lack of coincident measurements at sufficient spatial resolution, coordinating observa-88 tions and modeling are often crucial for understanding O_3 production from fire emissions Fiore, Pierce, Dickerson, Lin, and Bradley (2014). The diagnostics of trace-gas tendencies in 90 CTMs output are widely used to identify the drivers for ozone production due to var-91 ied anthropogenic and natural sources while fewer attempts for biomass burning sources 92 occur Barth et al. (2012); Hu, Xue, Kong, and Zhang (2019); Lu et al. (2018); Pfister 93 et al. (2019). In this study, the O_3 -tendency diagnostics, together with sensitivity sim-٩đ ulations both with and without fire emissions, allow exploration of the roles of chemi-95 cal and dynamical processes affecting vertical O_3 accumulation downstream of fires. The 96 enhancement of local vertical O_3 due to fire emissions is expected to arise from multi-97 ple processes, including the local chemical reactions (e.g., photochemical reaction of in-98 put O_3 precursors from fire emissions), transportation (e.g., the transported higher O_3 99 produced by upwind smoke plumes), and the vertical exchange (e.g., redistribution of 100 O_3 by interactions between surface and upper air). We are interested in understanding 101 the impact of those processes on vertical O_3 variability in fire smoke. 102

With relatively flat topography in the SEUS region, Huntsville station was usually 103 dominated by local anthropogenic/biogenic/agricultural burning at the surface and more 104 frequent wildfire plumes in the free troposphere (FT) Reid et al. (2017). In this case study, 105 we observed smoke affecting both surface and upper air over Huntsville downstream of 106 the wildfires. Using comprehensive observations (Huntsville ground-based lidars, in-situ 107 measurements, satellite) to evaluate the performance of regional model simulations re-108 sults in a tool to estimate the vertical variability of fire impacts. Integrating vertical ob-109 servations and modeling can benefit the evaluation work and scientific understanding. 110 The following objectives comprise this study: (1) Evaluate the model performance against 111 regional and local observations, especially vertical ozone and UV aerosol extinction pro-112 files and understand the model capabilities and limitations to reproduce the observations. 113 (2) Characterize the chemical and dynamical processes affecting the vertical ozone ac-114 cumulation in smoke plumes and understand the roles of local chemical reactions, trans-115 portation, and vertical exchange. (3) Quantify vertically the contribution from fire emis-116 sions to net chemical ozone production and particulate matter. 117

¹¹⁸ 2 Data and Methods

2.1 Wildfires Episode and Study Area

The 2016 Southeastern United States (SEUS) Wildfires series occurred along the 120 Southern Appalachians throughout October and November 2016 and burned over 158,000 121 acress across six Southern states (see Table S1 in the supporting information). Multiple 122 factors contributed to the extraordinary wildfires outbreak and spread, including the ex-123 ceptional drought, deep leaf litter and duff layers, many human ignitions with few light-124 ning strikes, episodic strong winds by frontal systems, and complex mountain landscapes 125 Konrad and Knox (2017). We focus on a high-pollution episode when smoke influenced 126 Huntsville station and the SEUS region in 12-14 November 2016. Figure 1a shows the 127 locations and names of 14 largest active wildfires around this study period. Figure 1b 128 shows the study domain with surface PM2.5 and O_3 monitoring sites. Huntsville sta-129 tion is located in North Alabama, with ground-based lidars and ozonesondes to be in-130 troduced in the following sections. 131

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2.2 Huntsville Station Facilities

Both the Ceilometer and the ground-based O_3 DIfferential Absorption Lidar (DIAL) at the UAH campus (34.725°N, 86.645°W) detect the vertical aerosol structure. Balloonborne Electrochemical Concentration Cell (ECC) ozonesonde launched from the UAH campus measures vertical O_3 concentration in smoke plume. Model simulations with both DIAL (aerosol extinction and O_3) and ozonesonde data assess how well the model captures the vertical distribution of O_3 and aerosol.

Although primarily designed for the detection of cloud heights, ceilometers have 139 the potential capability for a quantitative retrieval of aerosol backscatter coefficient Wieg-140 ner et al. (2014). The Vaisala CL51 ceilometer used in this study is a pulsed diode-laser 141 lidar (905 nm) in the UAH Mobile Integrated Profiling System (MIPS) Wingo and Knupp 142 (2015). For this case, the ceilometer, located on the UAH campus, measures backscat-143 ter profiles up to 15 km above ground level (AGL) with high spatial and temporal res-144 olution at 30 m and 15 s, respectively. Because the backscatter signal is dominated by 145 the aerosol component at 905 nm, the total backscatter intensity serves as an indicator of relative aerosol loading during 12-14 November 2016. 147

The UAH campus also houses one of the TOLNet O_3 DIAL systems, named the 148 Rocket-city O_3 Quality Evaluation in the Troposphere (RO₃QET) lidar. RO₃QET mea-149 sures vertical O_3 profiles from 0.1 km up to 10 km above the ground using 289 and 299-150 nm lasers with an uncertainty of about $\pm 10\%$ Kuang et al. (2011). The temporal res-151 olution of the lidar sampling is adjustable and is typically set at 10 minutes. The vertical resolution varies with altitude to obtain sufficient lidar signal-to-noise ratio and is 153 between 150 and 300 m in the planetary boundary layer (PBL). Aerosol extinction co-154 efficients at the non-absorption line (299 nm) are retrieved by assuming a constant aerosol 155 extinction-to-backscatter ratio, which is 60 sr for this study. Validation experiments through 156 comparing co-located high spectral resolution lidar (HSRL) observations suggest that 157 the RO_3QET lidar is capable of capturing aerosol variability at high spatio-temporal res-158 olution up to 6 km Kuang et al. (2020). 159

The Huntsville ECC ozonesonde attached with radiosonde provides vertical profiles of ozone, temperature, relative humidity (RH), and wind. In this study, the data derive from one of the weekly flights, which make observations from the surface up to 35 km with a vertical resolution of 100 m Newchurch et al. (2003) at the Huntsville ozonesonde station on the UAH campus. Measurements have precision better than $\pm 5\%$ and an accuracy better than $\pm 10\%$ for O₃ B. J. Johnson et al. (2002).

¹⁶⁶ 2.3 Surface Data and Satellite Products

Hourly PM2.5 and O_3 measurements retrieved from the Environmental Protection 167 Agency (EPA) (https://www.epa.gov/outdoor-air-quality-data) are used to evaluate the 168 model performance on surface air quality within the smoke-impacted region. Moderate 169 Resolution Imaging Spectroradiometer (MODIS) Collection 6 Level 2 10 km aerosol op-170 tical depth (AOD) data onboard Terra and Aqua (MOD04 L2 and MYD04 L2) Levy, 171 Hsu, et al. (2015) acquired from the NASA Earth Data Level-1 and Atmospheric Archive 172 & Distribution System Distributed Active Archive Center (LAADS DAAC) website (https://ladsweb.modaps.eosdis.n 173 evaluate the model performance on horizontal plume extent. Supporting information (Text 174 S1) details the AOD estimates from MODIS and the model. Visual images from MODIS 175 and the Visible Infrared Imaging Radiometer Suite (VIIRS), available via NASA World-176 view website (https://worldview.earthdata.nasa.gov/), qualitatively assess plume cov-177 erage and fire detections from fires and thermal anomalies products. The sensor reso-178

¹⁷⁹ lutions of MODIS and VIIRS hotspot detections are 1 km and 375 m, respectively.

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2.4 Model Description and Experiment Design

A fully coupled meteorology-chemistry model, the Weather Research and Forecast-181 ing with Chemistry model (WRF-Chem V3.9.1) is applied in this study. The model con-182 figurations are listed in Table 1. For this study we selected the Model for Ozone and Re-183 lated chemical Tracers (MOZART) gas phase chemical scheme Emmons et al. (2010) cou-184 pled with the Georgia Institute of Technology–Goddard Global Ozone Chemistry Aerosol 185 Radiation and Transport (GOCART) aerosol scheme Chin, Rood, Lin, Müller, and Thomp-186 son (2000), referred to as MOZCART Pfister et al. (2011). Other parameterizations in-187 clude Morrison's microphysics scheme, the Rapid Radiative Transfer Model (RRTM) long-188 wave and Goddard shortwave radiation schemes, the Monin-Obukhov surface layer, the 189 Noah Land Surface Model, the Yongsei University (YSU) PBL, the New Grell cumulus 190 scheme (G3), and the simplified Tropospheric Ultraviolet-Visible photolysis scheme (F-191 TUV). National Centers for Environmental Prediction (NCEP) North American Mesoscale 192 (NAM) 12 km Analysis data (https://rda.ucar.edu/datasets/ds609.0/, accessed 7 Feb 193 2018) provide the initial and lateral boundary meteorological conditions. MOZART-4 194 global model outputs provide the initial and lateral chemical conditions. Biomass burn-195 ing emissions are calculated using the Fire Inventory from NCAR (FINNv1.5) Wiedin-196 myer et al. (2011) and the online plume-rise model Freitas et al. (2007). FINNv1.5 are 197 based on fire counts derived from the Moderate Resolution Imaging Spectroradiometer (MODIS). The hourly emissions are allocated using the standard WRAP diurnal pro-199 file WRAP (2005). Anthropogenic emissions for both area and point sources are obtained 200 from the 2011 U.S. EPA national emissions inventory (NEI 2011 v2). Biogenic emissions 201 are calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) module Guenther et al. (2006). 203

Two nested domains cover CONUS and SEUS with 16 km and 4 km horizontal res-204 olutions, respectively. The vertical coordinate comprises 60 unequally spaced layers be-205 low 50 hPa, with 12 layers below 2 km altitude and a center height of 28 m for the low-206 est layer (see vertical grids structure in Figure S1). The simulation time period ranges 207 from 8 to 14 November 2016, for which the 12-14 November period in the inner domain 208 serves to avoid the influence of the model spinup during the first 4 days. The modeled 209 meteorology is reinitialized with analysis fields every 24 hours but the chemistry is re-210 cycled from the previous day. Three simulations estimate the wildfire impacts (Table 1): 211 simulation CTRL contains no fire emissions; simulation FIREorig contains the original 212 fire emissions (speciated from FINNv1.5 PM2.5) without emissions correction; simula-213 tion FIREcorr contains the fire emissions with emissions adjustment (description in Sec-214 tion 3.2). In order to generate identical meteorology for the sensitivity analysis on fire-215 impacted O_3 , the aerosol-radiation feedback is disabled. 216

217 2.5 Fire Inventories and Burn Area Products

The Monitoring Trends in Burned Severity database (MTBS; https://www.mtbs.gov) 218 provides input for total burn acres since ignition for sorting large wildfires (Table S1). 219 To investigate the emission inputs, the daily burn area is grouped for each wildfire by 220 aggregating FINNv1.5 burn area in the geospatial bounding box from MTBS wildfire database. 221 The fire emission inputs for this work (FIREorig and FIREcorr runs) are compared with 222 three MODIS-based fire inventories: the FINNv1.5, the Global Fire Assimilation Sys-223 tem version 1.2 (GFASv1.2) Kaiser et al. (2012), and the Quick Fire Emissions Database 224 version 2.5 r1 (QFEDv2.5 r1) Darmenov and da Silva (2013). 225

226 3 Results

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3.1 Horizontal and Vertical Plume Transport

In this section, analysis of the horizontal and vertical plume transport using satellite and ground-based lidars identifies the smoke-impacted period for model evaluation and diagnostic analysis later. This analysis also reveals that the daily and diurnal variations of smoke transport are mediated by synoptic weather and PBL evolution.

In Figure 2, MODIS AOD shows that the wildfires along the Southern Appalachi-232 ans continued to burn and emitted a significant amount of smoke over SEUS region in 233 12-14 November 2016. Thus, we select 12-14 November to compare the model with MODIS 234 AOD and surface monitors in later sections. NOAA WPC surface analysis (Figure S2) 235 shows that a cold front passed over the wildfire region during 12-18 LT on 11 Novem-236 ber. After the frontal passage, smoke stretched across portions of SEUS region by north-237 easterly wind on 12 November. As high-pressure circulation dominated the following two 238 days, AOD shows less spreading but more concentrated pattern around the wildfires. 239

The UAH ceilometer captured the aerosol plumes downwind of the fires, as shown 240 in Figure 3. The time-height curtain of backscatter intensity shows that several plumes 241 passed over Huntsville in the nighttime residual layer (RL) (Figure 3a). Some plumes 242 subsided toward surface before sunrise; others were entrained by a developing PBL in 243 the morning and then mixed vertically throughout the PBL. This mixing provides a mech-244 anism for fire emissions to contribute to the downwind air quality over night. In addi-245 tion, an elevated aerosol plume stayed at ~ 2 km AGL from 12 UTC (6 LT) on 12 Novem-246 ber to 12 UTC (6 LT) on 13 November, and it is likely to be fire smoke as MODIS shows 247 obvious smoke spreading over Huntsville. 248

The most severe surface particulate air pollution at Huntsville occurred on 13 Novem-249 ber, when an air-quality alert was issued for Madison and Morgan counties in the after-250 noon instigating DIAL measurement from 19:37 to 22:17 LT on 13 November under this 251 high aerosol loading condition. The time-height curtain of aerosol extinction coefficient 252 at 299 nm (Figure 3b) shows heavy background aerosols and relatively dense plumes within 253 that domain. The background aerosols below the capping inversion layer ($\sim 0.5/\text{km}$ be-254 low 1.5 km AGL) results from sufficient daytime mixing in the well-developed PBL. A 255 relatively dense plume (>1.0/km) features about 4 times higher extinction than that in 256 usual aerosol loading conditions. At 22 LT, the dense plume extended across the whole 257 RL column and evolved to be a thicker layer (~ 0.7 km thickness) between two finer lay-258 ers. The specified fine structure is highly consistent with that observed by ceilometer backscat-259 ter (Figure 3c). Ozonesonde and DIAL are used to assess the model performance on 12 260 and 13 November, respectively. 261

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3.2 Model Performance for AOD and Emissions Adjustment

In this section, the assessment of the model capability for reproducing the spatial pattern of smoke plumes as compared to satellite observations results in an adjustment of emission inputs. We justify this adjustment by comparing our emission inputs with three fire inventories, which give different emission estimations.

Comparing WRF-Chem AOD to MODIS AOD in 12-14 November (see details in 267 Text S2 and Figure S3) suggests that the model is able to reproduce the overall spatial 268 pattern of smoke plumes over SEUS, with a spatial correlation coefficient between mod-269 eled and observed AOD 0.6 on average. However, the model underestimated the AOD 270 magnitude. The domain-averaged observed AOD is about 3.6 times of the simulated AOD. 271 The cause could be uncertainties in emission estimations Pereira et al. (2016); Zhang et 272 al. (2014), inadequate assumptions of aerosol optical properties Curci et al. (2015), the 273 use of simplified aerosol chemistry modules without secondary organic aerosol Fast et 274 al. (2006), or misrepresentation of transport processes Aouizerats, Van Der Werf, Bal-275 asubramanian, and Betha (2015); Wu et al. (2017). In this case, the bias in predicting 276 the frontal passage could cause uncertainties to the smoke transport on 12 November (Fig-277 ure S2), and the aerosol scheme GOCART could also lead to uncertainties because it does 278 not include secondary organic aerosols. However, quantifying each bias is a challenge be-279 yond our scope. In this case study, we focus on exploring the uncertainties of emissions 280 only and increase the original fire aerosol emissions (PM2.5, PM10, organic carbon, black 281 carbon, and sulfate are speciated from PM2.5 in FINNv1.5) by a factor of 3.6, without 282 changing the fire gas-phase emissions. Even though this approach does not rectify all the 283 uncertainties in the emission estimates and may not reflect the temporal-spatial variations of smoke behavior, such a sensitivity study can help in constraining the emission 285 estimates based on satellite observations. 286

To justify this method for scaling the aerosol emissions, we compare the original 287 simulation (FIREorig) and the simulation with scaled aerosol emissions (FIREcorr) with 288 the inter-inventory differences. Figure 4 shows the ratio of carbonaceous aerosol (both 289 organic carbon OC and black carbon BC) and carbon monoxide CO in different fire in-290 ventories and our simulations. Each data point indicates the daily fire emission from the 291 fire inventories or our model inputs, summed over the wildfire region of interest defined 292 in 33.46-38.17°N and 78.75-86.25°W (see the selected region in Figure S4). The three 293 inventories show considerable discrepancies in the emissions ratios in November 2016 over 294 the wildfire area. The ratio is about 0.07, 0.09, and 0.4 in Gg/Gg for FINNv1.5, GFASv1.2, 295 and QFEDv2.5 r1, respectively. Such a broad range of emission ratios justifies our choice 296 to scale the emission input from 0.08 Gg/Gg to 0.3 Gg/Gg. 297

The discrepancies among fire inventories might arise from their different estima-298 tion processes. Although all of the three inventories are based on MODIS fire detections, 299 FINNv1.5 turns MODIS fire counts into burned area based on some assumptions Wied-300 inmyer et al. (2011), GFASv1.5 assimilates MODIS Fire Radiative Power (FRP) Kaiser 301 et al. (2012), and QFEDv2.5 r1 uses MODIS FRP directly combined with a scaling fac-302 tor by a top-down constraint for different biomes Darmenov and da Silva (2013). Carter 303 et al. (2020) showed that fire aerosol emissions from different inventories differ by a fac-304 tor of 4 to 7 over North America. Liu et al. (2020) showed that temperate North Amer-305 ica (TENA) has a coefficient of variation as high as 102% for mean annual OC+BC emis-306 sions among fire inventories. Our comparisons agree with previous assessment about the 307 uncertainties in fire aerosol emissions. The (OC+BC)/CO ratios over the SEUS wild-308 fire region differ by a factor of 5.7, within the discrepancy envelope of previous studies. 309 Because the performance of an individual fire inventory also depends on the region and 310 season, emission evaluation and adjustment is often necessary for a given fire event. 311

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3.3 Model Performance for Surface PM2.5 and Vertical Extinction

Using the adjusted emission inventory, we assess how well the model can simulate surface PM2.5 and vertical aerosol loading through comparing the model results with observations by surface monitors, lidar, and satellite.

Figure 5 shows the time series of U.S. EPA PM2.5 and modeled PM2.5 in three 316 sensitivity simulations. The FIREorig simulation shows obvious underestimation of ob-317 served PM2.5. Quantitatively, the standard deviations normalized with respect to ob-318 servations are much lower than 1.0 (see pattern statistics in Figure S5). After the emis-319 sion adjustment, the FIREcorr simulation is able to capture the domain-averaged mag-320 nitude and reproduce the maximum hourly PM2.5 ($\sim 200 \ \mu g/m^3$ on 12 November at Site 321 1). Additionally, the improved model reveals dominant fire contributions to the observed 322 PM2.5 exceedance of air quality standard (35 $\mu g/m^3$ for 24-hour limit), especially at the 323 rural sites nearby wildfires. Therefore, it is reasonable to use the FIREcorr simulation 324 to investigate the fire impacts on PM2.5. 325

Despite the improvement on magnitude, a domain-averaged scaling factor cannot 326 improve the model performance on the diurnal variations. Both FIREorig and FIREcorr 327 simulations perform well in reproducing the diurnal cycle at Site 3-7 but poorly at Site 328 1, 2, and 8. Statistically, modeled and observed PM2.5 have a fairly strong to moder-329 ate correlation at Site 3-7 and weak correlation at Site 1, 2, and 8. This model bias in 330 the diurnal fire behavior can be partly explained by satellites providing information at 331 the overpass time only Wang et al. (2006), wind bias, and the domain-averaged scaling 332 factor adopted here. Other potential error sources are discussed later. 333

Figure 6 shows the comparison between the DIAL-retrieved aerosol extinction (at 334 299 nm) and the simulated vertical aerosol extinction (at 300 nm) during 19-23 LT on 335 13 November. The FIREcorr simulation is able to capture the nocturnal boundary layer 336 aerosol (~ 0.5 /km below 1.5 km AGL), while the FIREorig simulation underestimated 337 the magnitude. This comparison indicates that the improved simulation can reproduce 338 the well-mixed smoke during the daytime PBL development. However, the FIREcorr sim-339 ulation missed the densest plume (>1.0/km after 20 LT on 13 November) observed by 340 DIAL, and it underestimated the highest MODIS AOD nearby wildfires (observed at noon 341 on 13 November in Figure S3) and PM2.5 at individual site nearby wildfire (e.g., Site 342 2). A likely reason for the underestimate is missing fire sources. 343

To confirm this hypothesis, we examine both MODIS and NPP/VIIRS reflectance 344 images with the fires and thermal anomalies product (Figure 7), and group the FINNv1.5 345 daily burn area by each wildfire (Figure 8). It was cloudy and hazy over the wildfires region on 13 November. Although some wildfires emitted visible dense smoke plumes (e.g., 347 the Rough Bridge Fire in north Georgia) and were counted in the NPP/VIIRS night de-348 tection, the fires were not counted in MODIS by the abnormal thermal product. Sub-349 sequently, the burn areas of many wildfires are zero on 13 November in FINNv1.5 inventory. Two other MODIS-based inventories (GFASv1.2 and QFEDv2.5 r1) also have quite 351 small aerosol and gas emissions on 13 November (Figure S7). These differences imply 352 that the clouds and thick haze probably obscured the MODIS fire detection on 13 Novem-353 ber. The detection limitation is probably associated with attenuated fire signal and so-354 lar heating during the day and the potential cloud/smoke classification issues Justice et 355 al. (2002); Polivka, Wang, Ellison, Hyer, and Ichoku (2016). As a result, the model us-356 ing the MODIS-based fire inventories could not reproduce some freshly-emitted smoke 357 plumes. 358

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3.4 Model Performance for Surface Ozone and Vertical Ozone

This section reports the modeled O₃ compared with surface monitors, ozonesonde, and DIAL measurements. Because the aerosol-radiation feedback has been turned off to generate identical meteorology, and MOZCART does not consider heteorgenous or aqueous chemistry, the modeled O₃ results in FIREorig and FIREcorr simulations are identical.

Figure 9 shows the time series of EPA O_3 and modeled O_3 in sensitivity simulations. Our model reproduced the observed surface O_3 level (below 60 ppb) during 12-

14 November at most sites. The results show consistent diurnal variations between sim-367 ulated and observed O_3 at both rural and urban sites, with strong to moderate corre-368 lation coefficients (see pattern statistics in Figure S6). The model performance at Site 369 3 and 5 is weaker, indicated by a low correlation coefficient and high normalized root-370 mean-square (NRMS) error. The model bias on O_3 can be complicated by meteorology, 371 emissions, and model parameterizations. In this case, uncertainty in fire emissions is not 372 the only possible source for the surface O_3 bias. Other factors, such as transport bias 373 during the frontal passage, model capability in reproducing nocturnal stable layer, and 374 the accuracy of anthropogenic and biogenic gaseous emissions, might also induce larger 375 model bias. The difference between FIREcorr and CTRL simulations suggests that the 376 total fire impacts on surface O_3 concentration was smaller than 10 ppb at most sites. 377

An ozonesonde launched from UAH campus at 13 LT on 12 November. Figure 10 378 compares modeled results with the observed vertical profiles, including ozone volume mix-379 ing ratio, relative humidity, potential temperature (θ) , horizontal wind speed, and hor-380 izontal wind direction. The ozonesonde reveals an enhanced O_3 lamina between two θ 381 inversion layers between 1.4-2.3 km. It peaks at 1.8 km AGL with 56 ppb, \sim 12 ppb larger 382 than PBL. This thick lamina co-existed with the elevated aerosol plume observed in ceilome-383 ter in light northeasterly wind. This coexistence of fire-impacted aerosol plume and en-384 hanced O_3 suggests a fire-impacted ozone lamina above the PBL height ~1.4 km. Over-385 all, WRF-Chem is able to reproduce vertical ozone and meteorological profiles in smoke 38 below 3 km. In particular, the model reproduces the wet and ozone-rich lamina, and ob-387 tains temperature and wind field consistent with observations; however, it is limited in 388 simulating finer inversion layers. The model predicts a lower PBL height of 1.2 km than 380 the observed 1.4 km, and it does not resolve the upper θ inversion at 2.3 km well. This 390 limitation is likely due to the relatively coarse vertical resolution at ~ 2 km and the bias 391 in predicting wind shear when the wind turned sharply above the lamina, as observed 392 by sonde. The underestimated O_3 in PBL is consistent with the underestimated surface 303 O_3 at the nearby site. This underestimate can be due partly to the model bias in wind 394 direction and relative humidity in PBL, as well as to other factors discussed. The model 395 also reproduces the O_3 laminae observed by DIAL during 19-23 LT on 13 November (Fig-396 ure 6), but it underestimates the O_3 magnitude in the nocturnal boundary layer, which 397 underestimate might be caused by the uncertainties in emission inputs as discussed in 398 the previous section. 399

400

3.5 Diagnosing Fire Impacts on Vertical O_3 and PM2.5 Distribution

Because the model performs well in simulating the vertical and surface ozone distributions and reproducing the well-mixed aerosol during daytime, the model calculations estimate the vertical ozone accumulation in fire smoke during daytime on 12 and 13 November 2016. We begin with a regional sensitivity analysis to show the overall fire impacts and the possible sources, and then apply the model's tendency diagnostics to further examine the processes contributing to the ozone accumulation over Huntsville. Fire-impacted PM2.5 indicates the altitude and strength of the fire smoke.

408

3.5.1 Regional Impacts at 13 LT on 12 and 13 November 2016

Figure 11 shows the modeled longitude-altitude curtain plots of O_3 , fire-impacted 409 O₃, and fire-impacted PM2.5 at 13 LT (19 UTC) on 12 November over the SEUS region. 410 The curtain shows that an enhanced O_3 lamina at 2 km ASL spreads widely from 78° W 411 to 88° W and passes over the Huntsville station (Figure 11a). This thick layer is spread-412 ing above PBL and is capped below 3 km by a strong wind shear when the wind turns 413 strongly westerly above ~ 3 km. Figure 11b and 11c show the modeled fire impacts (FIREcorr 414 minus CTRL) on O_3 and PM2.5, respectively. The simulations show enhanced O_3 con-415 centration within the elevated smoke plume, which are consistent with our observation 416 analysis. Quantitatively, the fire emissions result in an O_3 enhancement of 2-5 ppb and 417

PM2.5 enhancement of 10-20 $\mu g/m^3$ at 13 LT. In 85-86° W, the enhancements in O₃ and 418 PM2.5 can be much larger than 5 ppb and 20 $\mu g/m^3$, respectively. Using the modeled 419 hourly PM2.5 and AOD (not shown here), we estimate that the smoke plume is trans-420 ported to Huntsville from multiple wildfires that occurred during the frontal passage on 421 11 November (see the large active wildfires in Figure 8 and wildfire map in Figure 1a). 422 The wind-shear structure caps the mixed smoke plume with enhanced O_3 in the lowest 423 level of FT. Below the elevated plume, there is slightly lower PBL O_3 enhancement (1-424 2 ppb) and PM2.5 enhancement (5-15 $\mu g/m^3$) in 86-88° W. The smoke in the PBL is 425 relatively fresh with < 6 hrs transport time and is likely emitted from nearby small fire 426 on 12 November. 427

As the weather condition turns to high-pressure circulation on 13 November, a new 428 pattern emerges with concentrated fire impacts from the surface up to 2 km ASL on a 429 regional scale (Figure 12). PBL O_3 increases in the stagnant air, and fire emissions con-430 tribute more to O_3 and PM2.5. Quantitatively, the fire emissions result in a dominant 431 O_3 enhancement by 4-10 ppb or higher and PM2.5 enhancement by 40-80 μ g/m³ or higher 432 at 13 LT (Figure 12b and 12c). A large portion of the well-mixed PBL smoke is emit-433 ted from 12 November, when the wildfires are most active during our study period (Fig-434 ure 8). As illustrated in the observation analysis, the smoke remains in the residual layer 435 overnight and can effectively be transported to affect other locations on the next day. 436

437

3.5.2 Local Impacts in 7-17 LT on 12 and 13 November 2016

The sensitivity simulations confirm that fire emissions impacted the vertical ozone 438 contribution over Huntsville on 12 and 13 November. This local enhancement could be 439 caused by the transport of ozone and/or its precursors from fire emissions. The mod-440 eled results also imply that fire is not an exclusive source contributing to the observed 441 ozone laminae. This result brings up two questions: (1) What are the relative roles of 442 chemical and dynamical processes on the vertical ozone accumulation? (2) What is the 443 relative contribution of fire emissions to the total net photochemical ozone production? 444 To address these questions, we analyze the processes affecting vertical ozone distribu-445 tion through WRF-Chem tendency diagnostics, including net chemical ozone produc-446 tion PO₃ (Chem), horizontal and vertical advection of ozone (AdvH+AdvZ), and ver-447 tical mixing of ozone (Vmix). The daytime ozone tendency output from the sensitivity 448 simulations with fire emissions (FIREcorr) and without fire emissions (CTRL) is used 449 to explore the fire contribution. The following model results are averaged over 5×5 hor-450 izontal grids ($20 \text{ km} \times 20 \text{ km}$) over Huntsville for a better representativeness. 451

Figure 13a and 13b show daytime-integrated (7-17 LT) O_3 process tendencies and 452 PM2.5 over Huntsville in the FIREcorr and CTRL simulations on 12 and 13 November, 453 respectively. The absolute O_3 process tendencies show similar patterns on both days. In 454 the upper air (0.2-2.0 km AGL), the positive PO_3 dominates the daytime ozone accu-455 mulation on both days. The total PO_3 peaks on 13 November in the middle PBL by up 456 to 19 ppb/10 hrs at 0.5 km. In the surface layer below 0.2 km, pronounced negative PO_3 457 is caused by the quick NOx titration near the surface (modeled NOx ~ 15 ppb). The neg-458 ative PO_3 is offset by positive O_3 contributions from vertical mixing and advection pro-459 cesses. Vertical mixing contributes positively near the surface yet negatively in the up-460 per air, because it tends to disperse the enhanced O_3 from the upper air to the surface Hu 461 et al. (2019). The results imply that local chemical processes dominates the upper air 462 ozone accumulation while dynamical processes directly contribute to the build-up of ground-463 level ozone over Huntsville. 464

Figure 13c and 13d extract the relative O₃ tendencies and PM2.5 contributed by
fire emissions (FIREcorr minus CTRL) on 12 and 13 November, respectively. Here FIREcorrCTRL SumTend (SumTend means the sum of all process tendencies) indicates the daytimeintegrated ozone change due to fire emissions (Figure S8). During 12-13 November, fire

emissions increase the vertical O_3 concentrations by affecting local chemical reactions, 469 transportation, and the vertical exchange. The total daytime ozone increase due to fire 470 emissions is similar on both days, with largest contributions at lower altitude (up to ~ 8 471 ppb). However, the dominant processes contributing to the total signals show daily and 472 vertical variability. On 12 November, an increase of positive PO_3 dominates the upper-473 level (above 1.2 km) fire-impacted O_3 accumulation, while the transport process dom-474 inates at the lower level. In contrast, on 13 November, an increase in PO_3 (either through 475 increased ozone chemical production or a decrease in ozone chemical loss) dominates the 476 lower level (below 1.0 km), while the transport processes dominate at the upper level. 477 The decrease of negative PO_3 in the surface layer (i.e., PO_3 is more negative in CTRL 478 compared to FIREcorr below 0.2 km) is affected by the additional NOx and VOCs from 479 the fire emissions. 480

Quantitatively, the percentage contribution from fire emissions to the net chem-481 ical ozone production is calculated by (FIREcorr-CTRL)/FIREcorr results during day-482 time over Huntsville. Fire emissions contribute 14% to the highest daytime PO₃ on 12 483 November (2 ppb out of 17 ppb at 1.6 km) and 25% on 13 November (5 ppb out of 19 18/ ppb at 0.5 km). This different photochemical production is associated with the varied 485 fire emissions and smoke transport. The smoke strength is indicated by the fire-impacted 486 PM2.5 here. The percentage contribution from fire emissions to vertical hourly PM2.5 487 peaks at 51% on 12 November (10 μ g/m³ out of 19 μ g/m³ at 1.8 km) and 77% on 13 November (37 $\mu g/m^3$ out of 48 $\mu g/m^3$ at 1.0 km). The results suggest an increased fire 489 contribution to the enhancement of ozone and particulate matter from day to day. 490

Diurnal variability of process tendencies can be affected by the boundary layer evo-491 492 lution, transport changes over the course of the day, and photochemistry. To examine how the different processes vary over the day, we analyze the total and fire-impacted pro-493 cess tendencies for 7-9 LT, 11-13 LT, and 15-17 LT in Figure 14. The total PO_3 clearly 494 peaks in the middle of the day. The total advection term dominates in the late afternoon 495 on 12 November and the middle of the day on 13 November when the largest inflow of 496 ozone occurred. The total vertical mixing process is strongest when the PBL is built up 497 in the middle of the day, and it dominates the surface ozone accumulation by dispers-498 ing considerable upper air ozone downward. 499

The diurnal variability of the total tendency terms can help explain what processes 500 drive the ozone increase from fire emissions during different times of the day. On 12 Novem-501 ber, transport process in the late afternoon drives the largest fire impacts on O_3 accu-502 mulation (~4 ppb/2 hrs near the surface), which is associated with the freshly emitted smoke plume discussed. Fire-impacted PO_3 from morning to the middle of the day is 504 not intensive, yet this local chemical reaction dominates the ozone increase in the ele-505 vated smoke plume (compared to little dynamical process tendencies). On 13 Novem-506 ber, the middle of the day drives the largest ozone increase (> 4 ppb/2 hrs), with high-507 est PO_3 and inflow of O_3 from fire emissions, as well as the strongest vertical mixing that 508 dispersing O_3 to the lower level. The combined effect of chemical reaction, transport, 509 and vertical mixing on O_3 accumulation is pronounced in the middle of the day when 510 fire smoke impacted the boundary layer. 511

512 4 Conclusions

This case study for the 2016 Southeastern Wildfires illustrates the high spatio-temporal variations of smoke impacts on air quality at both regional and local scales. Integrating vertical aerosol and O₃ profiles with modeling and multi-platform observations can fill the knowledge gap at altitudes above ground to improve our understanding of the fire impacts on vertical aerosol and O₃ distribution. In particular, the fire-impacted processes and fire-source contributions on 12 and 13 November are demonstrated by sensitivity simulations and tendency diagnostics: (1) Fire emissions increase the vertical ozone concen-

trations downstream of the fires by affecting local net chemical ozone production, inflow 520 and outflow of ozone, and vertical ozone exchange. The dominant process has daily, di-521 urnal, and vertical variability due to the PBL evolution, photochemistry, and smoke trans-522 port changes over the course of the day. On 12 November, local photochemical ozone pro-523 duction over Huntsville dominates the fire-impacted ozone enhancement in the elevated 524 plume; while transport process dominates the boundary layer ozone accumulation in the 525 late afternoon. (2) In this study, biomass burning is not a dominant source contribut-526 ing to the local chemical ozone production (positive PO_3 is increased by up to 25% in 527 the upper air), but can still play an important role in changing the O_3 concentrations 528 because of additional impacts of vertical mixing and advection processes. The combined 529 effect of chemical and dynamical processes lead to an increase of O_3 concentration by 530 up to ~ 8 ppb during daytime at lower altitude. In the upper air, the increased concen-531 tration is smaller than 8 ppb, but the relative contribution from fire emissions to the to-532 tal O_3 increase can be dominant. Fire emissions contribute significantly to the vertical 533 accumulation of PM2.5 (by up to 77%) during daytime. Besides the freshly emitted smoke 534 plumes, relatively aged plumes emitted from previous day contribute considerably to the 535 local PM2.5 accumulation. 536

In this case, WRF-Chem can capture the general day-to-day AOD pattern, air qual-537 ity variations, vertical structure of aged plumes, and enhanced ozone lamina. Three main 538 avenues for future work: (1) Discrepancies in fire emission estimations need to be con-539 sidered for model inputs. (OC+BC)/CO emission ratios in FINNv1.5, GFASv1.2, and 540 QFEDv2.5 r1 fire inventories differ by a factor of 5.7 (in Gg per Gg) over the 2016 SEUS 541 wildfire regions. A scaling ratio of 3.6 on aerosol emissions (derived from FINNv1.5 PM2.5), 542 within the spanned range of the emission ratios in different inventories, can improve the 543 modeled magnitude of surface PM2.5, vertical aerosol extinction, and AOD, but this was 544 tested in only a single case study. (2) After the emission adjustment, underestimation 545 of the densest plume in DIAL and highest AOD in MODIS is partly due to missing fire 546 detections under clouds on 13 November. Adding extra satellite detections (e.g., FINNv2.2 547 includes VIIRS information) or filling in the gap of missing fire counts in emission es-548 timation algorithms could be considered; (3) The density, continuity, and species of ver-549 tical measurements are relatively limited for modeling evaluation in this case study. Com-550 bining larger samples of vertical measurements (ground-based and airborne) with am-551 bient data will benefit regional-model evaluation in future fire studies. 552

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- data is acquired from the NASA Earth Data Level-1 and Atmospheric Archive & Dis-
- tribution System Distributed Active Archive Center (LAADS DAAC) website (https://ladsweb.modaps.eosdis.nasa.go
- The WRF-Chem preprocessor tools and emission inputs used in this study are available
- via https://www2.acom.ucar.edu/wrf-chem/wrf-chem-tools-community.

Simulations	1. CTRL (fire off) 2. FIREorig (fire on) 3. FIREcorr (fire on, correction)
Vertical	60 vertical levels from the surface to 50 hPa (vertical grids in Figure S1)
Horizontal	D01: 16 km×16 km, D02: 4 km×4 km
Emissions	Fire: FINNv1.5, Anthropogenic: NEI 2011 v2, Biogenic: MEGAN
IC&BC	Met: NAM 12 km, Chemical: MOZART global
Chemistry	MOZART gas, GOCART aerosol
Physics	Goddard, RRTM, Morrison's, Monin-Obukhov, Noah, YSU, G3, F-TUV

\mathbf{Fab}	le 1.	Key	Configuration	is for the	WRF-Chem	v3.9.1	Simulations
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Figure 1. (a) Map of 14 active wildfires (red triangles) during 11-14 November, 2016 (see
Table S1 for details). Here the names of wildfires are defined by the Monitoring Trends in Burn
Severity project (MTBS; www.mtbs.gov). (b) WRF-Chem inner domain (D02) and terrain height
(m). Black dots, blue dots, and magenta dot represent the 8 EPA PM2.5 sites, 6 EPA O₃ sites,
and Huntsville station, respectively. The magenta line indicates the cross section of D02 across
Huntsville station used for Figure 11.



Figure 2. MODIS aerosol optical depth (AOD) at 550 nm in SEUS region on 12, 13, and 14
November 2016, respectively. MODIS AOD is calculated by mean of Aqua AOD at 19 UTC (13
LT) and Terra AOD at 17 UTC (11 LT) (or 16 UTC (10 LT) on 13 November). Cross marker
indicates Huntsville location.



Figure 3. (a) Time-height curtain of UAH CT25K ceilometer backscatter intensity in 12-14
November 2016 (courtesy of Kevin Knupp). Here UTC time minus 6 hours is local time. The
black triangle indicates the launch time of an ozonesonde. The black rectangle indicates the measurement time of DIAL. (b) Time-height curtain of DIAL aerosol extinction coefficient at 299 nm
in 1:37-4:17 UTC on 14 November (19:37 to 22:17 LT 13 November). (c) Same time period with
(b), but for ceilometer backscatter intensity at 905 nm.



Figure 4. Comparison of fire emission ratios for (OC+BC) versus CO between this work
and the different inventories. Daily emissions in the wildfire region are summed up within the
latitude and longitude boundary 33.46-38.17° N and 78.75-86.25° W (Figure S4). The black,
red, and blue dots represent daily emissions in 1-30 November 2016 from FINNv1.5, GFASv1.2,
and QFEDv2.5_r1 inventories, respectively. Unfilled aqua and orange squares represent daily
emissions in 8-14 November 2016 from FIREorig and FIREcorr runs, respectively. The gray dots
denote a scaling by 3.6 on the original FINNv1.5 aerosols for a reference.



Figure 5. Comparison between 8 EPA sites (black line) and WRF-Chem hourly PM2.5 in 1214 November 2016 for control run CTRL (aqua), before the emissions adjustment FIREorig (red),
and after the emissions adjustment FIREcorr (brown). The control run is performed to show the
modeled PM2.5 without fire impacts. Pattern statistic can be seen in Figure S5. 8 EPA PM2.5
sites include: 1. Asheville, NC, 2. Mitchell, NC, 3. Swain, NC, 4. Greenville-Anderson-Mauldin,
SC, 5. Chattanooga, TN-GA, 6. Nashville-Davidson-Murfreesboro-Franklin, TN, 7. Macon, GA,
8. Decatur, AL.



Figure 6. Modeled time-height curtain of aerosol extinction coefficient (at 300 nm) and ozone
before (FIREorig) and after (FIREcorr) the emissions adjustment in 1-5 UTC on November 14
(19-23 LT on 13 November), compared with DIAL aerosol extinction (at 299 nm) and ozone.



Figure 7. Corrected Reflectance, Fires and Thermal Anomalies on 13 November 2016 from
(upper) Terra and Aqua/MODIS (Day and Night); (middle) NPP/VIIRS (Day, 375 m); (bottom)
NPP/VIIRS (Night, 375 m). Image source: NASA Worldview.



Figure 8. FINNv1.5 daily burn area (acres) group by individual wildfire during 11-14 November 2016. The daily burn area is aggregated in the geospatial boundary box of each wildfire that defined by MTBS database.



Figure 9. Comparison between EPA (black line) and WRF-Chem ozone in 12-14 November
2016 for control run (light blue), before correction (red), and after correction (brown). Pattern
statistic can be seen in Figure S6. 6 EPA O₃ sites include: 1. Great Smoky Mountains NP-Look
Rock, TN, 2. Cranberry, NC, 3. Sand Mountain, AL, 4. St.Andrews State Park, Panama City
Beach, FL, 5. Coweeta, NC, 6. South DeKalb, GA.

-28-



Figure 10. Comparison between ozonesonde (black color) and WRF-Chem FIREcorr simulation (red color) at 19 UTC (13 LT) on 12 November 2016. Ozone volume mixing ratio (O₃), relative humidity (RH), potential temperature (θ), horizontal wind speed (WS), and horizontal wind direction (WD) are displayed respectively. The aqua lines represent the PBL heights from sonde (solid line) and model (dashed line).



Figure 11. (a) Modeled (FIREcorr) vertical sections of O₃ mixing ratio (ppb) from west to
east in SEUS region across Huntsville latitude (34.72° N) at 19 UTC (13 LT) on 12 November
2016 at 0-4 km ASL altitude. Solid red line denotes the longitude of Huntsville. Arrows indicate
modeled direction and speed of horizontal wind. (b) Same as Figure a, but for fire-impacted
(FIREcorr minus CTRL) O₃ mixing ratio. Note the colorbar range is different from Figure a. (c)
Same as Figure b, but for fire-impacted PM2.5 concentration (µg/m³).





Figure 12. Same as Figure 11, but for 19 UTC (13 LT) on 13 November 2016.



Figure 13. (a) and (b) Process analysis of daytime-integrated vertical ozone tendencies and
daytime-averaged PM2.5 over Huntsville in simulations with (FIREcorr, solid lines) and without
(CTRL, dashed lines) fire emissions during 7-17 LT on 12 and 13 November, respectively. Processes include chemical reactions (Chem, red), horizontal and vertical advections (AdvH+AdvZ,
blue), vertical mixing (Vmix, gray), and summed tendencies of all processes (SumTend, black).
PM2.5 is represented by green lines. (c) and (d) are same as Figure (a) and (b) but for fire-

impacted values, calculated by the difference between FIREcorr and CTRL simulations.



Figure 14. Same as Figure 13, but present diurnal variability by integrating 2 hours in 7-9,
11-13, and 15-17 LT on 12 November (row 1, 3) and 13 November (row 2, 4), respectively.

study_area_v2.



modisAOD.



OCplusBCvsCO_Gg.



ceilometer_dial_v4.

(a) Ceilometer Backscatter in November 12-14, 2016



wrfchem_pm25.



ext_o3_evaluation_v2.



satellite_detection.



burn_area.



wrfchem_ozone_v2.



HU995_20161112_wofb.



alt_lon_nov12_wofb.



alt_lon_nov13_wofb.



daytime_tendency_v6.



2hr_tendency_v10.

