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## RESEARCH ARTICLE

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### Key Points:

- Fires contribute 12–32  $\mu\text{g}/\text{m}^3/\text{hr}$  (44%–70%) to surface PM<sub>2.5</sub> and 7–8 ppb/10 h (30%–37%) to daytime ozone at Huntsville
- Fire-impacted ozone below 1 km is dominated by advection on November 12 and by local photochemistry on November 13
- Increasing aerosol fire emissions by a factor of 3–4 better matches observed AOD, PM<sub>2.5</sub>, and DIAL aerosol extinction

### Supporting Information:

Supporting Information may be found in the online version of this article.

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## Impact of the 2016 Southeastern US Wildfires on the Vertical Distribution of Ozone and Aerosol at Huntsville, Alabama

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**Abstract** We present an integrated analysis of measurements from ozonesonde, ozone ( $\text{O}_3$ ) Differential Absorption Lidar (DIAL), ceilometer, surface monitors, and space-borne observations in conjunction with the regional chemical transport model Weather Research and Forecast Model with Chemistry (WRF-Chem) to investigate the effect of biomass burning emissions on the vertical distribution of ozone and aerosols during an episode of the 2016 Southeastern United States wildfires. The ceilometer and DIAL measurements capture the vertical extent of the smoke plumes affecting the surface and upper air over Huntsville, AL. The model evaluation results suggest a scaling factor of 3–4 for the wildfire aerosol emissions in order to better match observed aerosol optical depth, fine particulate matter (PM<sub>2.5</sub>), and DIAL aerosol extinction. We use the scaled emissions together with WRF-Chem tendency diagnostics to quantify the fire impacts and characterize the processes affecting the vertical ozone budget downstream of the wildfires. During the daytime at Huntsville on November 12 and 13, we estimate that fire emissions contribute 12–32  $\mu\text{g}/\text{m}^3$  (44%–70%) to hourly surface PM<sub>2.5</sub> and 7–8 ppb/10 h (30%–37%) to the surface ozone increase ( $\Delta\text{O}_3$ ), respectively. Net chemical ozone production ( $\text{PO}_3$ ) is the main contributor to upper-air ozone, which reaches 17–19 ppb/10 h with an estimated 14%–25% contribution from fire sources. Vertical mixing and advection are the major drivers of changes in surface ozone. Model analysis indicates that advection dominates  $\Delta\text{O}_3$  due to fire emissions below 1 km on November 12, while local photochemistry dominates on November 13. These results quantify the different mechanisms through which fires can influence the vertical ozone budget and point out uncertainties in fire inventories that need to be addressed in light of the increasing role of wildfires on air quality.

### 1. Introduction

Biomass burning (BB) releases substantial amounts of aerosol and ozone precursors that can affect climate and air quality (Akagi et al., 2011; Andreae & Merlet, 2001; Crutzen & Andreae, 1990; Crutzen et al., 1979). Previous observation and modeling studies have indicated that BB emissions contribute to local and regional air-quality problems (Baker et al., 2016; Hodzic et al., 2007; Jaffe et al., 2013; Pfister et al., 2008; Wigder et al., 2013), as well as to downwind pollution via long-range transport (Colarco et al., 2004; Cook et al., 2007; Jaffe et al., 2004; Lapina et al., 2006; Lindaas et al., 2017; Martin et al., 2006; McKeen et al., 2002; Morris et al., 2006; Oltmans et al., 2010; Rogers et al., 2020; Sapkota et al., 2005). These studies demonstrate that the impacts of biomass burning on air quality can vary dramatically over time and space. Models can be used to investigate the mechanisms through which BB influences air quality. However, more efforts are required to assess how current models capture the variability of BB impacts, especially at higher spatial and temporal resolution.

Chemical transport models (CTMs) have been widely used to estimate fire impacts. CTMs can provide good spatiotemporal coverage, differentiate the impacts of specific sources, and support mechanistic understanding of chemical and dynamical processes (Baker et al., 2018); however, amongst other issues, large uncertainties in fire-emission estimates present challenges for estimating the variability of fire impacts. These uncertainties can arise from limitations of satellite detection and inherent uncertainties of the empirical approaches used for emission estimations (Carter et al., 2020; Justice et al., 2002; Liu et al., 2020; van der Werf

et al., 2017; Wang et al., 2018). The emission factors (EFs, the mass of a pollutant emitted per unit mass of biomass burned) are critical inputs for fire-emission models, but they vary by fuel type and fire conditions. While knowledge of EFs has increased substantially over the past decade, the uncertainty and natural variation in EFs remained a large source of uncertainties in BB emission estimates (Akagi et al., 2011; Andreae & Merlet, 2001). Furthermore, a large discrepancy of emission estimates could exist between the bottom-up and top-down approaches. For instance, top-down constraints are applied on aerosol emissions to match smoke aerosol optical depth (AOD) (Darmenov & da Silva, 2013; Kaiser et al., 2012). However, these global constraints are not applied in all fire inventories and may bias smoke estimates at regional to local scales. Recent studies show that temperate North America has much larger discrepancies in carbonaceous aerosols estimates than many other regions (Carter et al., 2020; Liu et al., 2020). These studies highlight the importance of further investigation and require observations at all scales (surface, aloft, and satellite).

Although satellites and surface monitors make routine measurements of atmospheric O<sub>3</sub> concentration, balloon soundings and lidar techniques can provide precise vertically resolved O<sub>3</sub> observations throughout the troposphere and lower stratosphere (Thompson et al., 2011). This vertical information significantly benefits air-quality management and modeling improvement (Cooper et al., 2015). We take advantage of both ozonesonde (Newchurch et al., 2003) and ozone lidar (Kuang et al., 2011) techniques at the University of Alabama in Huntsville (UAH) to measure the vertical distribution of key atmospheric parameters. The UAH ozone lidar is affiliated with the Tropospheric Ozone Lidar Network (TOLNet, <https://www-air.larc.nasa.gov/missions/TOLNet/>). Under a collaborative protocol, the TOLNet lidars have demonstrated their feasibility and capability in fire studies (M. Johnson et al., 2016, 2021; Kuang et al., 2017; Langford et al., 2015, 2020; Reid et al., 2017; Strawbridge et al., 2018) and scientific projects (Gronoff et al., 2019; Leblanc et al., 2018; Sullivan et al., 2019). The continuous profiling of ozone and aerosols provides details missed by isolated measurements and is an asset for model evaluation by coordinating measurements (Langford et al., 2018, 2019). In addition, the ultraviolet (UV) backscatter (or extinction) profiles retrieved from ozone lidar can quantify the aerosol variability at the high spatiotemporal resolution, and these measurements serve as a tracer for fire smoke (Kuang et al., 2020; Langford et al., 2020). To our best knowledge, there has been little or no attempt to evaluate CTMs using this range-resolved UV aerosol optical product.

Coordinating vertical observations and simulations can also improve our understanding of the fire impacts on vertical profiles. Due to multiple O<sub>3</sub> sources in the troposphere and a lack of coincident measurements at sufficient spatial resolution, combining observations and modeling is crucial for understanding the O<sub>3</sub> production from fire emissions (Fiore et al., 2014). The diagnostics of trace-gas tendencies in CTMs output are widely used to identify the drivers for ozone production due to varied anthropogenic and natural sources (Barth et al., 2012; Hu et al., 2019; Lu et al., 2018; Pfister et al., 2019), while fewer studies for biomass burning sources have been completed. In this study, the O<sub>3</sub>-tendency diagnostics, together with sensitivity simulations both with and without fire emissions, allow exploration of the roles of chemical and dynamical processes affecting the vertical O<sub>3</sub> accumulation downstream of fires. The enhancement of the local vertical O<sub>3</sub> distribution due to fire emissions is expected to arise from multiple processes, including local photochemistry (e.g., photochemical reaction of the O<sub>3</sub> precursors from fire emissions), transport of ozone by upwind smoke plumes, and the vertical exchange (e.g., redistribution of O<sub>3</sub> by interactions between surface and upper air). In this study, we coordinate a range of different data sets and methods to understand the impact of these different processes on the vertical O<sub>3</sub> variability in fire smoke.

With a relatively flat topography in the Southeastern United States (SEUS) region, the region around Huntsville, AL was usually dominated by local anthropogenic, biogenic, and agricultural burning emissions at the surface, while wildfire plumes typically prevail in the free troposphere (FT) (Reid et al., 2017). Our case study of an episode when Huntsville was affected by the 2016 SEUS wildfires shows that smoke can affect ozone and aerosol loadings at the station both at the surface and in the upper air. Using comprehensive observations (Huntsville ground-based lidars, in situ measurements, space-borne observations) to evaluate the performance of regional model simulations results in a framework to estimate the vertical variability of these fire impacts. Coordinating vertical observations and modeling provides additional value in assessing model performance and enhances the scientific understanding. The following objectives comprise this study: (1) Evaluate the model performance against regional and local observations, especially vertical ozone and UV aerosol extinction profiles, to understand the model capabilities and limitations in reproducing the

observations. (2) Characterize the chemical and dynamical processes affecting the vertical ozone accumulation in smoke plumes and understand the roles of local chemical reactions, transportation, and vertical exchange. (3) Vertically quantify the contribution from fire emissions to net chemical ozone production and particulate matter.

## 2. Data and Methods

### 2.1. Wildfires Episode and Study Area

The 2016 SEUS wildfires series occurred along the Southern Appalachians throughout October and November 2016 and burned over 158,000 acres across six Southern states (see Table S1). Multiple factors contributed to the extraordinary wildfire outbreak and spread, including an exceptional drought, deep leaf litter and duff layers, many human ignitions with relatively few lightning strikes, episodic strong winds by frontal systems, and complex mountain landscapes (Konrad & Knox, 2017; Williams et al., 2017). We focus on a high-pollution episode when smoke influenced the Huntsville station and the SEUS region during November 12–14, 2016. Figure 1a shows the locations and names of the 14 largest active wildfires during this study period. Figure 1b shows the study domain with surface PM<sub>2.5</sub> and O<sub>3</sub> monitoring sites indicated. The Huntsville station is located in North Alabama on the UAH campus.

### 2.2. Huntsville Station Facilities

Both the ceilometer and the ground-based O<sub>3</sub> DIfferential Absorption Lidar (DIAL) at the UAH campus (34.725°N, 86.645°W) detect the vertical aerosol structure. Balloon-borne electrochemical concentration cell (ECC) ozonesondes launched from the UAH campus allow the measurement of the vertical O<sub>3</sub> concentration in smoke plumes. Both DIAL (aerosol extinction and O<sub>3</sub>) and ozonesonde data are used to assess how well the model captures the vertical distribution of O<sub>3</sub> and aerosol loadings.

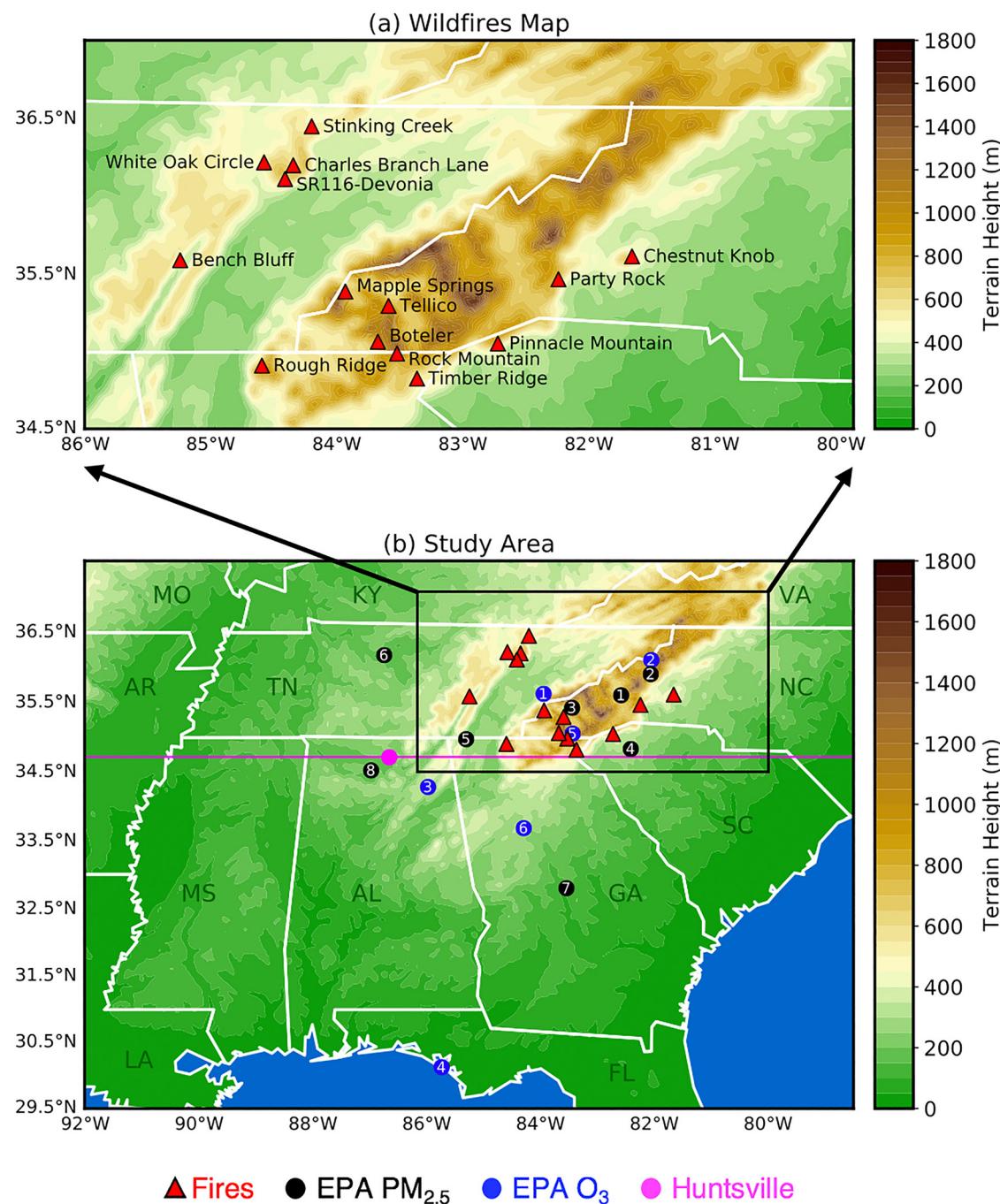
Although primarily designed to detect cloud heights, ceilometers have the capability for a quantitative retrieval of the aerosol backscatter coefficient (Wiegner et al., 2014). The Vaisala CL51 ceilometer used in this study is a pulsed diode-laser lidar (905 nm) in the UAH Mobile Integrated Profiling System (MIPS) (Wingo & Knupp, 2015). The ceilometer, located on the UAH campus, measures backscatter profiles up to 15 km above ground level (AGL) with high spatial and temporal resolution at 30 m and 15 s, respectively. Because the backscatter signal is dominated by the aerosol component at 905 nm, the total backscatter intensity serves as an indicator of relative aerosol loading during November 12–14, 2016.

The UAH campus also houses one of the TOLNet O<sub>3</sub> DIAL systems, named the Rocket-city O<sub>3</sub> Quality Evaluation in the Troposphere (RO<sub>3</sub>QET) lidar. RO<sub>3</sub>QET measures vertical O<sub>3</sub> profiles from 0.1 km up to 10 km above the ground using 289 and 299 nm lasers with an uncertainty of about ±10% (Kuang et al., 2011). The temporal resolution of the lidar sampling is adjustable and is typically set at 10 min. The vertical resolution varies with altitude to obtain a sufficient lidar signal-to-noise ratio and is between 150 and 300 m in the planetary boundary layer (PBL). Aerosol extinction coefficients at the non-absorption line (299 nm) are retrieved by assuming a constant aerosol extinction-to-backscatter ratio, which is 60 steradians (sr) for this study. Validation experiments through comparing with co-located high spectral resolution lidar observations suggest that the RO<sub>3</sub>QET lidar is capable of capturing aerosol variability up to 6 km altitude at high spatiotemporal resolution (Kuang et al., 2020).

The Huntsville ECC ozonesonde attached with a radiosonde provides vertical profiles of ozone, temperature, relative humidity (RH), and wind. In this study, the data are derived from one of the weekly flights at the Huntsville ozonesonde station, which makes observations from the surface up to 35 km with a vertical resolution of 100 m (Newchurch et al., 2003). The ozone measurements have a precision better than ±5% and an accuracy better than ±10% (B. Johnson et al., 2002).

### 2.3. Surface Data and Satellite Products

Hourly PM<sub>2.5</sub> and O<sub>3</sub> measurements retrieved from the Environmental Protection Agency (EPA) (<https://www.epa.gov/outdoor-air-quality-data>) are used to evaluate the model performance for the surface air



**Figure 1.** (a) Map of 14 active wildfires (red triangles) during November 11–14, 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](http://www.mtbs.gov)). (b) WRF-Chem inner domain (D02) and terrain height (m). Black dots, blue dots, and magenta dot represent the eight EPA PM<sub>2.5</sub> sites, six EPA O<sub>3</sub> sites, and Huntsville station, respectively. The magenta line in the inner model domain (D02) indicates the route of the vertical section in Figure 10.

quality within the smoke-impacted region. Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 6 Level 2 10 km merged Dark Target/Deep Blue AOD data onboard Terra and Aqua (MOD04\_L2 and MYD04\_L2) (Levy et al., 2015) acquired from the NASA Earth Data Level-1 and Atmospheric Archive & Distribution System Distributed Active Archive Center (LAADS DAAC) website (<https://ladsweb.modaps.eosdis.nasa.gov/>) is used to evaluate the model performance for the horizontal plume extent. To compare MODIS AOD at 10-km resolution with WRF-Chem AOD at 4-km resolution, we regrid both MODIS and

**Table 1***Key Configurations for the WRF-Chem v3.9.1 Simulations*

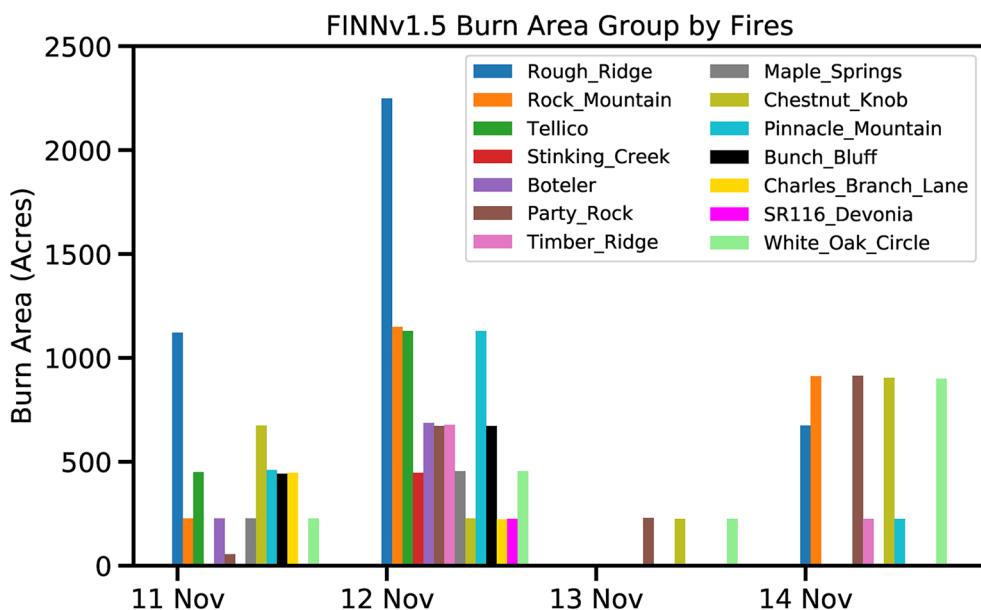
Simulations	1. CTRL (fire off)	2. FIREorig (fire on)	3. FIREcorr (fire on, correction)
Vertical	60 vertical levels from the surface to 50 hPa (vertical grids in Figure S1)		
Horizontal		D01: 16 × 16 km, D02: 4 × 4 km	
Emissions		Fire: FINNv1.5, Anthropogenic: NEI 2011 v2, Biogenic: MEGAN	
IC&BC		Met: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

modeled AOD to a  $0.1^\circ \times 0.1^\circ$  grid. For each grid box, MODIS AOD at 550 nm is calculated as the mean of Aqua AOD at 19:00 UTC and Terra AOD at 17:00 UTC (or 16:00 UTC on November 13), WRF-Chem AOD at 550 nm is calculated as the mean of modeled AOD at 19:00 UTC and modeled AOD at 17:00 UTC (or 16:00 UTC on November 13). Because WRF-Chem can directly output the variable “EXTCOF55,” which represents layer aerosol extinction coefficients for 550 nm, we define modeled AOD at 550 nm as the vertical sum of each the “EXTCOF55” field multiplied by the layer depth. Visual images from MODIS and the Visible Infrared Imaging Radiometer Suite (VIIRS), available via the NASA Worldview website (<https://worldview.earthdata.nasa.gov/>), are used to qualitatively assess smoke plume coverage and fire detection from thermal anomalies. The sensor resolutions of MODIS and VIIRS hotspot detections are 1 km and 375 m, respectively.

#### 2.4. Model Description and Experiment Design

A fully coupled meteorology-chemistry model, the Weather Research and Forecasting with Chemistry model WRF-Chem (Fast et al., 2006; Grell et al., 2005) version 3.9.1 is applied in this study. The model configurations are listed in Table 1. For this study, we selected the Model for Ozone and Related chemical Tracers (MOZART) gas-phase chemical scheme (Emmons et al., 2010) coupled with the Georgia Institute of Technology–Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) aerosol scheme (Chin et al., 2000), referred to as MOZCART (Pfister et al., 2011). Other parameterizations include the Morrison's microphysics scheme, the Rapid Radiative Transfer Model (RRTM) longwave and Goddard shortwave radiation schemes, the Monin-Obukhov surface layer, the Noah Land Surface Model, the Yonsei University (YSU) PBL, the New Grell cumulus scheme (G3), and the simplified Tropospheric Ultraviolet-Visible photolysis scheme (F-TUV). National Centers for Environmental Prediction North American Mesoscale (NAM) 12 km Analysis data (<https://rda.ucar.edu/datasets/ds609.0/>, accessed February 7, 2018) provide initial and lateral boundary meteorological conditions. MOZART-4 global model outputs provide the initial and lateral chemical conditions. Biomass burning emissions are calculated using the Fire Inventory from NCAR (FINNv1.5) (Wiedinmyer et al., 2011) and the online plume-rise model (Freitas et al., 2007). FINNv1.5 is based on fire counts derived from the MODIS. The hourly emissions are allocated using the standard WRAP diurnal profile (WRAP, 2005). For the MOZCART scheme, the aerosol emissions are speciated from FINNv1.5 particulate matter (PM). The speciation as provided in the emission preprocessor is listed in Table S2. Anthropogenic emissions for both area and point sources are obtained from the 2011 US EPA national emissions inventory (NEI 2011 v2). Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) module (Guenther et al., 2006).

Two nested domains cover CONUS and SEUS with 16 and 4 km horizontal resolutions, respectively. The vertical coordinate comprises 60 unequally spaced layers below 50 hPa, with 12 layers below 2 km altitude and a center height of 28 m for the lowest layer (see vertical grids structure in Figure S1). The simulation time period ranges from November 8 to 14, 2016 with the first 4 days used as model spin-up. To ensure that the model represents accurate meteorology, we use nudged meteorological fields and in addition conduct a meteorology reinitialization every 24-h, but recycle the initial chemical fields from the end of the previous day. The evaluation of the surface temperature is shown in Figure S11 and demonstrates the continuity of simulated meteorological fields despite the 24-h reinitialization. Three simulations are performed to



**Figure 2.** FINNv1.5 daily burn area (acres) group by individual wildfire during November 11–14, 2016. The daily burn area is aggregated in the geospatial boundary box of each wildfire that defined by the MTBS database.

estimate the wildfire impacts (Table 1): the CTRL simulation contains no fire emissions; the FIREorig simulation contains the original fire emissions (speciated from FINNv1.5 PM<sub>2.5</sub> and PM<sub>10</sub>) without emissions correction; the FIREcorr simulation contains the fire emissions with emissions adjustment (description in Section 3.2). To generate identical meteorology for the sensitivity analysis on fire-impacted O<sub>3</sub>, the aerosol-radiation feedback is disabled.

## 2.5. Fire Inventories and Burn Area Products

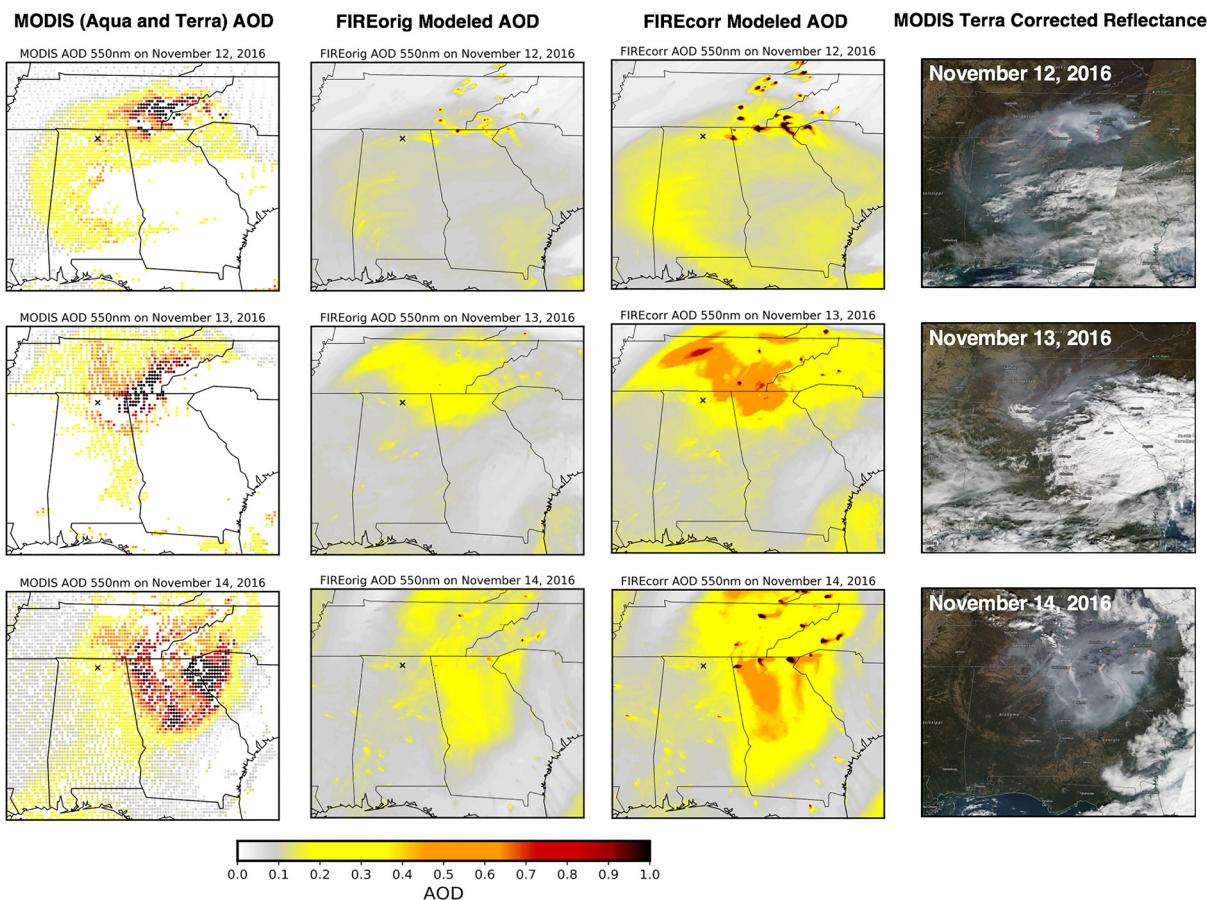
The Monitoring Trends in the Burned Severity database (MTBS; <https://www.mtbs.gov>) provides information on the total number of acres burned since ignition and is used to indicate large wildfires (Table S1). To investigate the emission inputs, we estimate the daily burn area for each wildfire (Figure 2) by aggregating the FINNv1.5 burn area in the geospatial bounding box from the MTBS wildfire database. The fire emission inputs for this work (FIREorig and FIREcorr simulations) are compared with four MODIS-based fire inventories: FINNv1.5, the Global Fire Emissions Database (GFEDv4s) (van der Werf et al., 2017), the Global Fire Assimilation System version 1.2 (GFASv1.2) (Kaiser et al., 2012), and the Quick Fire Emissions Database version 2.5\_r1 (QFEDv2.5\_r1) (Darmenov & da Silva, 2013).

## 3. Results

### 3.1. Horizontal and Vertical Plume Transport

In this section, we discuss the horizontal and vertical plume transport using satellite and ground-based lidars and identify the smoke-impacted period, which is used later for model evaluation and diagnostic analysis. Our analysis reveals that the daily and diurnal variations of smoke transport are mediated by synoptic weather conditions and the PBL evolution.

In Figure 3, MODIS AOD and MODIS imagery show that the wildfires along the Southern Appalachians continued to burn and emitted a significant amount of smoke over the SEUS region during November 12–14, 2016. This time period is chosen for later model evaluation and analysis. NOAA WPC surface analysis (Figure S2) shows that a cold front passed over the wildfire region during 12–18 LT on November 11. After the frontal passage, smoke stretched across portions of the SEUS region driven by northeasterly winds on

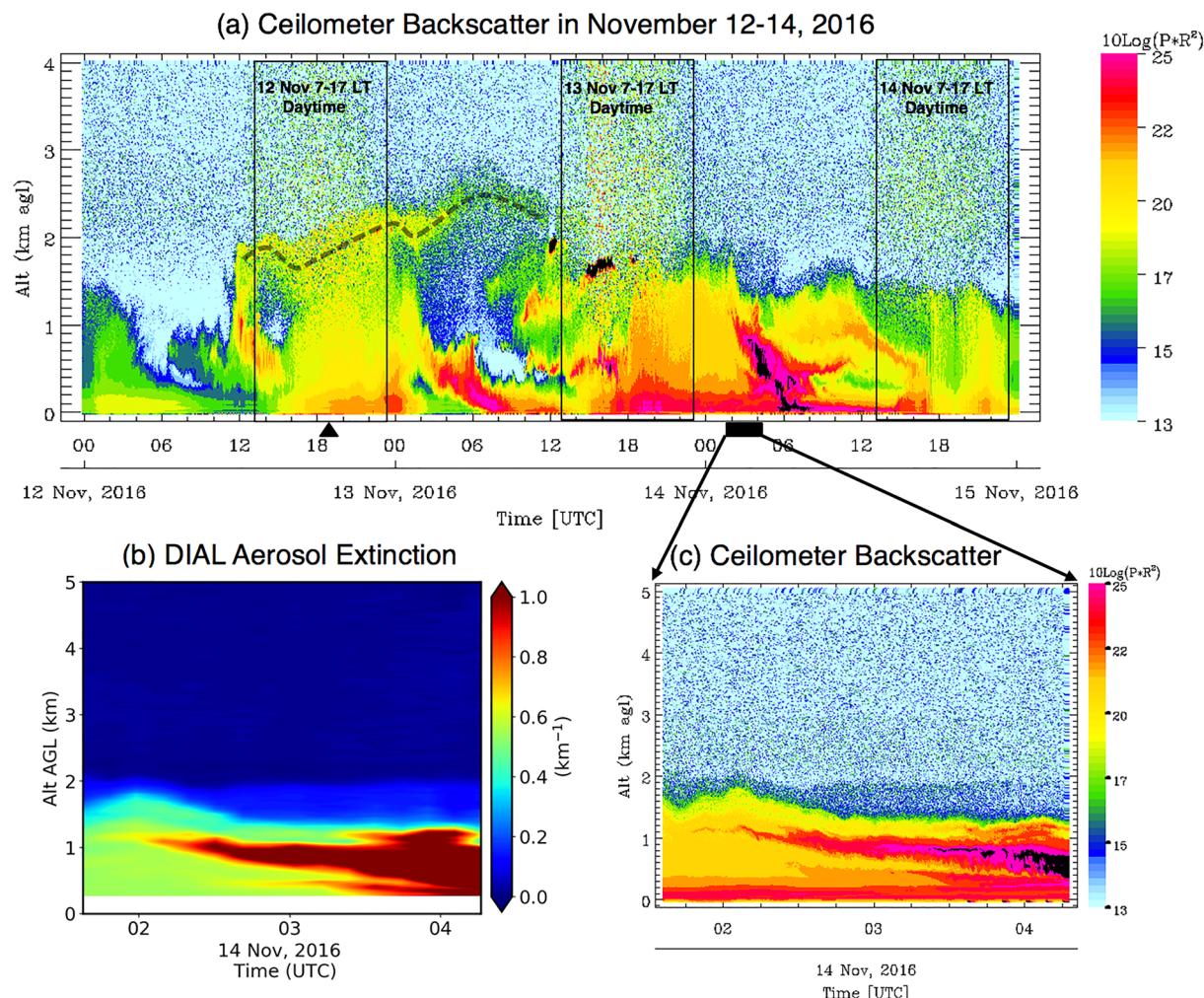


**Figure 3.** MODIS Versus WRF-Chem AOD at 550 nm in SEUS region in (Row 1–3) November 12–14, 2016. Columns 1–4 show MODIS AOD, FIREorig AOD (before adjustment), FIREcorr AOD (after adjustment), and MODIS reflectance, respectively. Cross marker indicates Huntsville location. In the MODIS reflectance images, the red dots represent the fires and thermal anomalies product.

November 12. As a high-pressure circulation dominated the following 2 days, AOD shows the less spreading pattern, with a more concentrated distribution around the location of the wildfires.

The UAH ceilometer captured the aerosol plumes downwind of the fires, as shown in Figure 4. The time-height curtain of backscatter intensity shows that several plumes passed over Huntsville in the nighttime residual layer (RL) (Figure 4a). Some plumes subsided toward the surface before sunrise; others were entrained by a developing PBL in the morning and then mixed vertically throughout the PBL. This mixing provides a mechanism for fire emissions to contribute to the downwind air quality overnight. In addition, an elevated aerosol plume stayed at ~2 km AGL from 12:00 UTC (06:00 LT) on November 12 to 12:00 UTC (06:00 LT) on November 13. This plume is likely fire smoke as MODIS shows obvious smoke spreading over Huntsville (Figure 3).

The most severe surface particulate air pollution at Huntsville occurred on November 13, when an air-quality alert was issued for Madison and Morgan counties in the afternoon instigating DIAL measurement from 19:37 to 22:17 LT on November 13 under this high aerosol loading condition. The time-height curtain of the aerosol extinction coefficient at 299 nm (Figure 4b) shows heavy background aerosols and relatively dense plumes within that domain. The background aerosols below the capping inversion layer (~0.5/km below 1.5 km AGL) result from sufficient daytime mixing in the well-developed PBL. A relatively dense plume (>1.0/km) features about 4 times higher extinction (>1.0/km) compared to typical aerosol loading conditions. At 22 LT, the dense plume extended across the whole RL column and evolved to be a thicker layer (~0.7 km thickness) between two finer layers. The specified fine structure is highly consistent with the



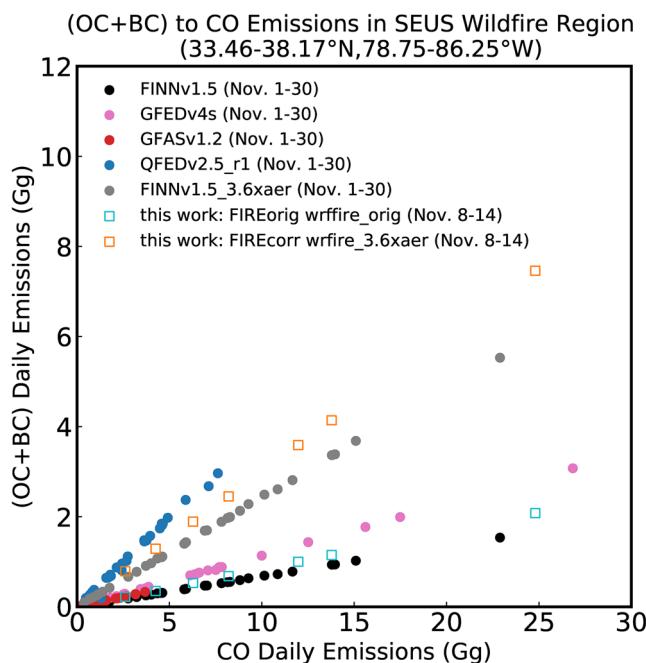
**Figure 4.** (a) Time-height curtain of UAH CL51 ceilometer backscatter intensity in November 12–14, 2016 (courtesy of Kevin Knupp). Here, UTC time minus 6 h is local time. The black triangle indicates the launch time of an ozonesonde. The black rectangle indicates the measurement time of DIAL. (b) Time-height curtain of DIAL aerosol extinction coefficient at 299 nm in 01:37–04:17 UTC on November 14 (19:37–22:17 LT November 13). (c) The same time period with (b), but for ceilometer backscatter intensity at 905 nm.

ceilometer backscatter observation (Figure 4c). Ozonesonde and DIAL measurements are used to assess the model performance on November 12 and 13, respectively.

### 3.2. Model Performance for AOD and Emissions Adjustment

In this section, the modeled spatial pattern of smoke plumes is assessed against satellite observations. Results uncover limitations that lead to an adjustment of the emission inputs, which is further justified by comparing our emission inputs with four fire inventories.

Comparing WRF-Chem AOD to MODIS AOD for November 12–14 (Figures 3 and S3) suggests that the simulation “FIREorig” is able to reproduce the overall spatial pattern of smoke plumes over the SEUS, with a spatial correlation coefficient between modeled and observed AOD of 0.6 on average (0.4 on November 12, 0.7 on November 13 and 14). The lower correlation on November 12 is associated with the prediction bias of the frontal passage during 00:00–18:00 UTC on November 12, indicated by the front locations in surface analysis and modeled result (Figure S2). Despite the general agreement in AOD distribution, there is an underestimation of the model in the AOD magnitude. We calculate an averaged scaling ratio  $r$  by averaging the three different slopes for November 12–14 (MODIS AOD/FIREorig Modeled AOD = 3.6).



**Figure 5.** 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°N–38.17°N and 78.75°W–86.25°W (Figure S4). The black, pink, red, and blue dots represent daily emissions in November 1–30, 2016 from FINNv1.5, GFEDv4s, GFASv1.2, and QFEDv2.5\_r1 inventories, respectively. Unfilled aqua and orange squares represent daily emissions in November 8–14, 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. Note the recommended scaling factor 3.4 on GFASv1.2 aerosol emissions by Kaiser et al. (2012) is not shown in this plot.

Thus, the domain-averaged observed AOD is about 3.6 times the simulated AOD. The reasons for this could be uncertainties in emission estimations (Pereira et al., 2016; Zhang et al., 2014), inadequate assumptions of aerosol optical properties (Curci et al., 2015), the use of simplified aerosol chemistry modules without representation of secondary organic aerosol (SOA) (Fast et al., 2006), or misrepresentation of transport processes (Aouizerats et al., 2015; Wu et al., 2017). Quantifying each bias is a challenge beyond our scope. In this case study, we focus on exploring the uncertainties of emissions only and increase the original fire aerosol emissions by a factor of 3.6, without changing the fire gas-phase emissions. Specifically, we multiply all aerosol components (unspeciated PM<sub>2.5</sub>, PM<sub>10</sub>, organic carbon, black carbon, and sulfate) in the original gridded fire emissions by 3.6. This adjustment improves the model performance of the domain-averaged AOD and changes the average slope from 3.6 (MODIS vs. FIREorig Modeled AOD) to 1.1 (MODIS vs. FIREcorr Modeled AOD), as shown in Figure S3. Although this approach does not rectify all the uncertainties in the emission estimates and may not reflect the temporal-spatial variations of smoke behavior, our sensitivity study can help constrain the emission estimates based on satellite observations.

To justify the scaling of the fire aerosol emissions, we compare emissions from the original simulation (FIREorig) and the simulation with scaled aerosol emissions (FIREcorr) to the inter-inventory differences. Figure 5 shows the ratio of carbonaceous aerosols (both organic carbon OC and black carbon BC) to carbon monoxide CO in different fire inventories and our simulations. Each data point indicates daily fire emission summed over the wildfire region of interest defined in 33.46°N–38.17°N and 78.75°W–86.25°W (see the selected region in Figure S4). The four inventories show considerable discrepancies in the emissions ratios for November 2016 over the wildfire area. The (OC + BC)/CO ratio is about 0.07, 0.09, 0.11 and 0.4 in Gg/Gg for FINNv1.5, GFASv1.2, GFEDv4s and QFEDv2.5\_r1, respectively. Such a broad range of emission ratios justifies our choice to scale the emission input from 0.08 Gg/Gg to 0.3 Gg/Gg.

Emission differences may arise from inventory-specific methods for estimating the amount of burned vegetation, the vegetation type, and the emission factors. Although all of the four inventories are based on MODIS fire detections, FINNv1.5 converts the MODIS active fire product into burned area generally by assuming an upper limit of area burned and further scaling it by the percent of bare cover (Wiedinmyer et al., 2011); GFEDv4s primarily uses the MODIS burn area product retrieved from pre-burn and post-burn images and ingests active fire locations for its small fire boost (Giglio et al., 2013; Randerson et al., 2012; van der Werf et al., 2017); GFASv1.5 assimilates MODIS Fire Radiative Power (FRP) (Kaiser et al., 2012); QFEDv2.5\_r1 uses MODIS FRP directly combined with a scaling factor derived by a top-down constraint for different biomes (Darmenov & da Silva, 2013). All four inventories also differ in the vegetation types used, and while they generally use emission factors from Andreae and Merlet (2001) and Akagi et al. (2011), they use different updates and also aggregate these emission factors differently. Consequently, the (OC + BC)/CO ratios over the SEUS wildfire region differ by a factor of 5.7 (Figure 5). Our analysis is consistent with the previous assessment about the uncertainties in fire aerosol emissions. Carter et al. (2020) showed that fire aerosol emissions from different inventories differ by a factor of 4–7 over North America. Liu et al. (2020) showed that temperate North America has a coefficient of variation as high as 102% for mean annual OC + BC emissions among fire inventories. Our choice of a scaling factor of 3.6 sets our emissions within this discrepancy envelope. The scaling of fire aerosol emissions is also supported by previous studies (Darmenov & da Silva, 2013; Kaiser et al., 2012; Wiedinmyer et al., 2011), which addressed the need for observation constraints on the bottom-up estimates of fire aerosol emissions, especially for a given fire event.

### 3.3. Model Performance for Surface PM<sub>2.5</sub> and Vertical Extinction

Using the adjusted emission inventory, we assess how well the model simulates surface PM<sub>2.5</sub> and the vertical aerosol loading by comparing the model results with observations from surface monitors, lidar, and satellite.

Figure 6 shows the time series of US EPA PM<sub>2.5</sub> observed and modeled PM<sub>2.5</sub> from three sensitivity simulations. The FIREorig simulation shows an obvious underestimation of observed PM<sub>2.5</sub> with a domain-wide mean bias of  $-21.6 \mu\text{g}/\text{m}^3$  against hourly observations from all eight sites. The standard deviations normalized with respect to observations are much lower than 1.0 (see pattern statistics in Figure S5), with a domain-averaged value of 0.3 among all eight sites. After the emission adjustment, the FIREcorr simulation is able to capture the domain-averaged magnitude (as the mean bias reaches  $-0.4 \mu\text{g}/\text{m}^3$  and the normalized standard deviation reaches 1.0) and reproduce the maximum hourly PM<sub>2.5</sub> ( $\sim 200 \mu\text{g}/\text{m}^3$  on November 12 at Site 1). In addition, the FIREcorr simulation reveals dominant fire contributions to the observed PM<sub>2.5</sub> exceedance of the air quality standard ( $35 \mu\text{g}/\text{m}^3$  for 24-h limit), especially at the rural sites nearby the wildfires.

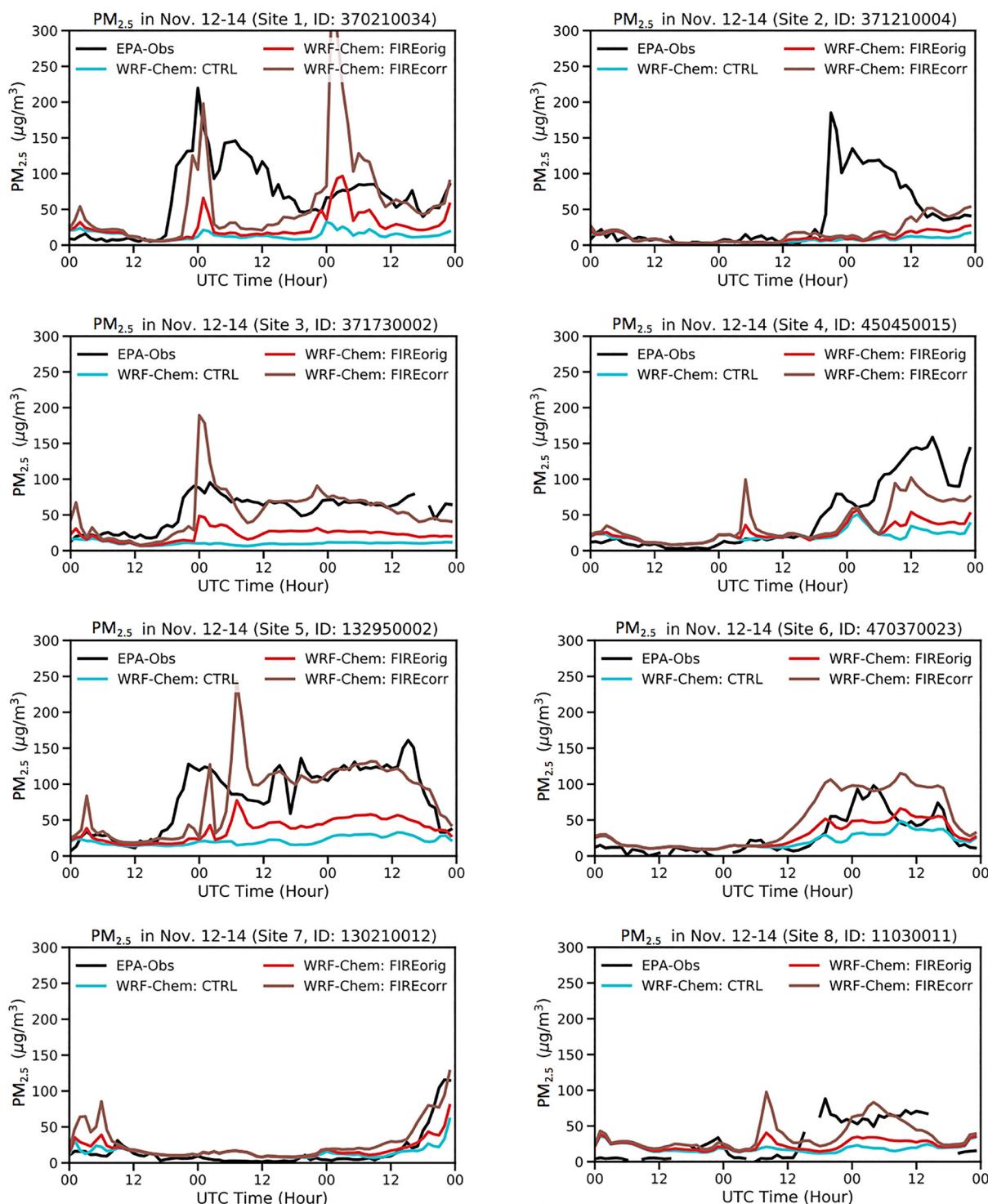
Despite the improvement in magnitude, the domain-averaged correlation coefficient just slightly increases from 0.5 to 0.6, as a domain-averaged scaling factor cannot improve the model performance for the diurnal variations. Both FIREorig and FIREcorr simulations perform well in reproducing the diurnal cycle at Sites 3–7 but perform poorly at Sites 1, 2, and 8. Statistically, modeled and observed PM<sub>2.5</sub> have a strong to moderate correlation at Sites 3–7 and weak correlation at Sites 1, 2, and 8. This model bias in the diurnal fire behavior can be partly explained by satellites providing information at the overpass time only (Wang et al., 2006), biases in the wind field, and the domain-averaged scaling factor adopted here. Other potential error sources are discussed later.

Figure 7 shows the comparison between the DIAL-retrieved aerosol extinction (at 299 nm) and the simulated vertical aerosol extinction (at 300 nm) for 19:00–23:00 LT on November 13. The FIREcorr simulation is able to capture the nocturnal boundary layer aerosol extinction ( $\sim 0.5/\text{km}$  below 1.5 km AGL), while the FIREorig simulation underestimated the magnitude. This comparison indicates that the improved simulation can reproduce the well-mixed smoke during the daytime PBL development. However, the FIREcorr simulation misses the densest plume ( $>1.0/\text{km}$  after 20:00 LT on November 13) observed by DIAL, underestimates the highest MODIS AOD nearby wildfires (observed at noon on November 13 in Figure 3), and underestimates PM<sub>2.5</sub> at one site nearby the wildfire (e.g., Site 2). A likely reason for the underestimate is missing fire sources.

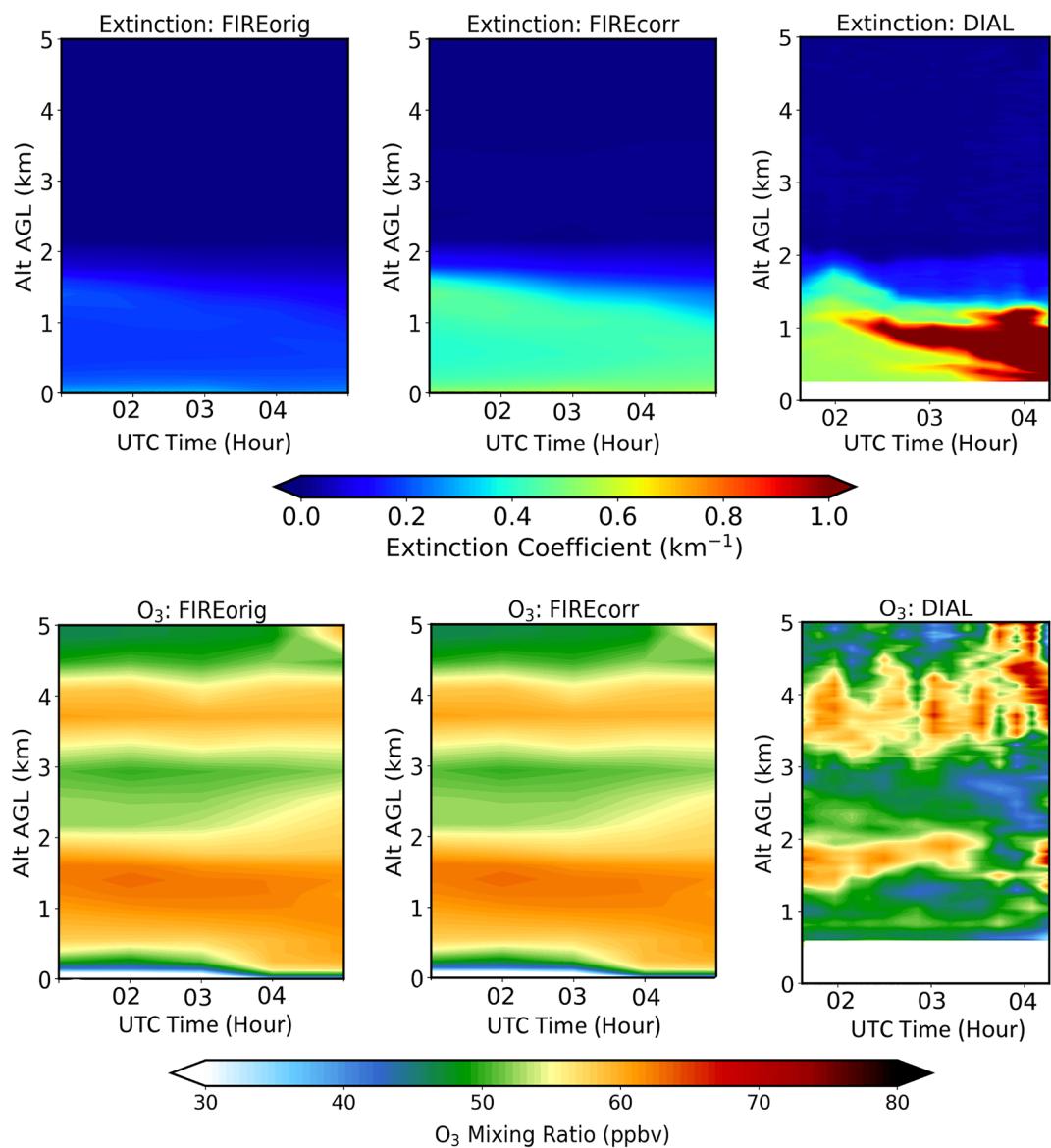
To confirm this hypothesis, we examine both MODIS and NPP/VIIRS reflectance images with the fires and thermal anomalies product (Figure S9), compared to the FINNv1.5 daily burn area grouped by each wildfire (Figure 2). It was cloudy and hazy over the wildfires region on November 13, conditions that obscure satellite detection. Although some wildfires emitted visible dense smoke plumes (e.g., the Rough Bridge Fire in north Georgia) and were counted in the NPP/VIIRS night detection, the fires were not counted by the MODIS thermal anomalies product. Consequently, the burn areas for many wildfires are zero on November 13 in the FINNv1.5 inventory. Two other MODIS-based inventories (GFASv1.2 and QFEDv2.5\_r1) also report small aerosol and gas emissions on November 13 (Figure S7). These differences imply that the clouds and thick haze probably obscured the MODIS fire detection on November 13. The detection limitation is likely associated with attenuated fire signal and solar heating during the day and the potential cloud/smoke classification issues (Justice et al., 2002; Polivka et al., 2016). As a result, the model using the MODIS-based fire inventories could not reproduce some freshly emitted smoke plumes.

### 3.4. Model Performance for Surface Ozone and Vertical Ozone

This section reports the comparison of modeled O<sub>3</sub> with surface monitors, ozonesonde, and DIAL measurements. Because aerosol-radiation feedback has been turned off in the simulations to generate identical meteorology, and heterogenous or aqueous chemistry is not considered in the MOZCART mechanism, the modeled O<sub>3</sub> is identical in the FIREorig and FIREcorr simulations.

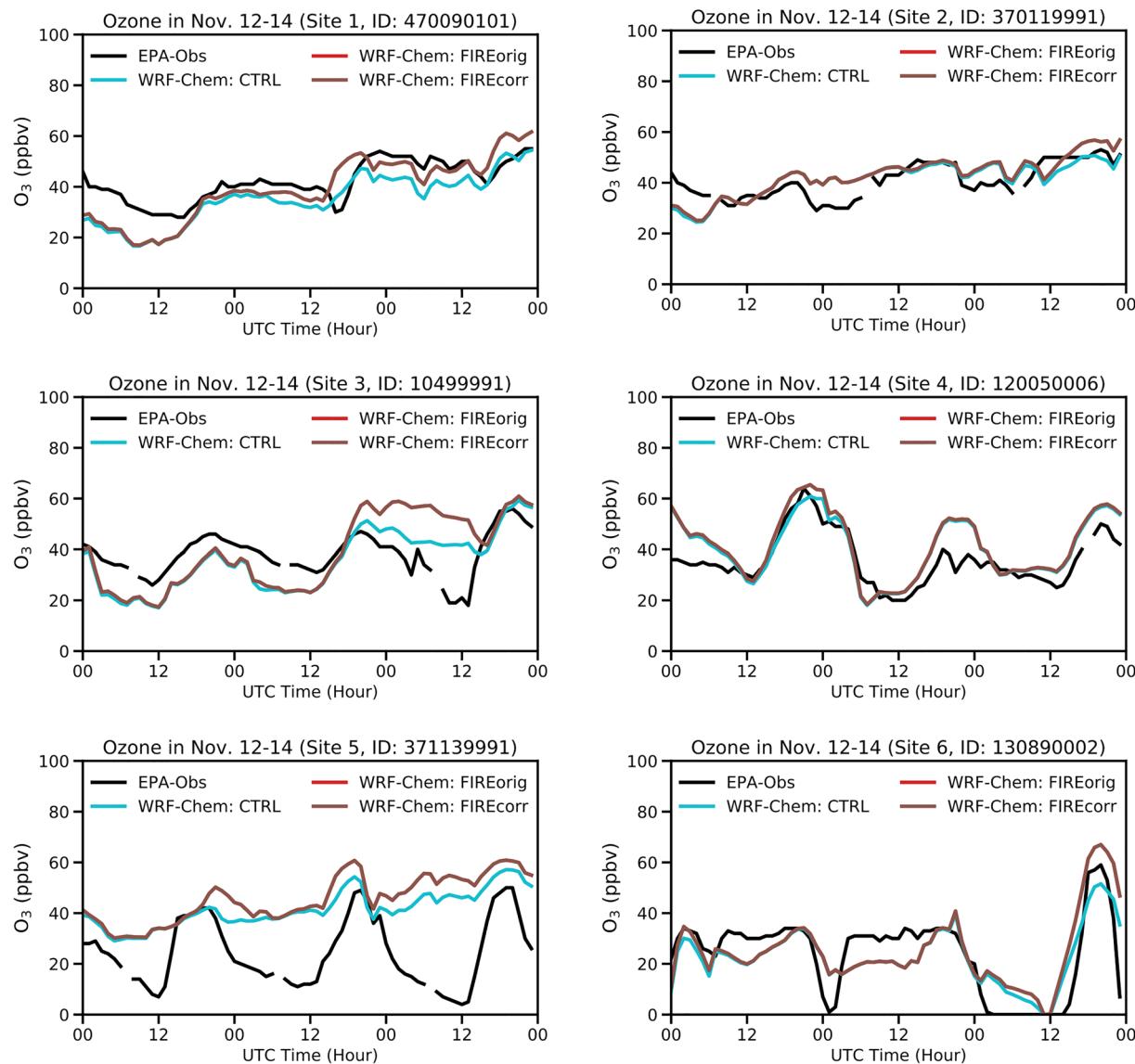


**Figure 6.** Comparison between eight EPA sites (black line) and WRF-Chem hourly PM<sub>2.5</sub> in November 12–14, 2016 for the control run CTRL (aqua), before the emissions adjustment FIREorig (red), and after the emissions adjustment FIREcorr (brown). The control run is performed to show the modeled PM<sub>2.5</sub> without fire impacts. Pattern statistic can be seen in Figure S5. Eight EPA PM<sub>2.5</sub> 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 7.** Modeled time-height curtain of aerosol extinction coefficient (at 300 nm) and ozone before (FIREorig) and after (FIREcorr) the emissions adjustment in 01:00–05:00 UTC on November 14 (19:00–23:00 LT on November 13), compared with DIAL aerosol extinction (at 299 nm) and ozone.

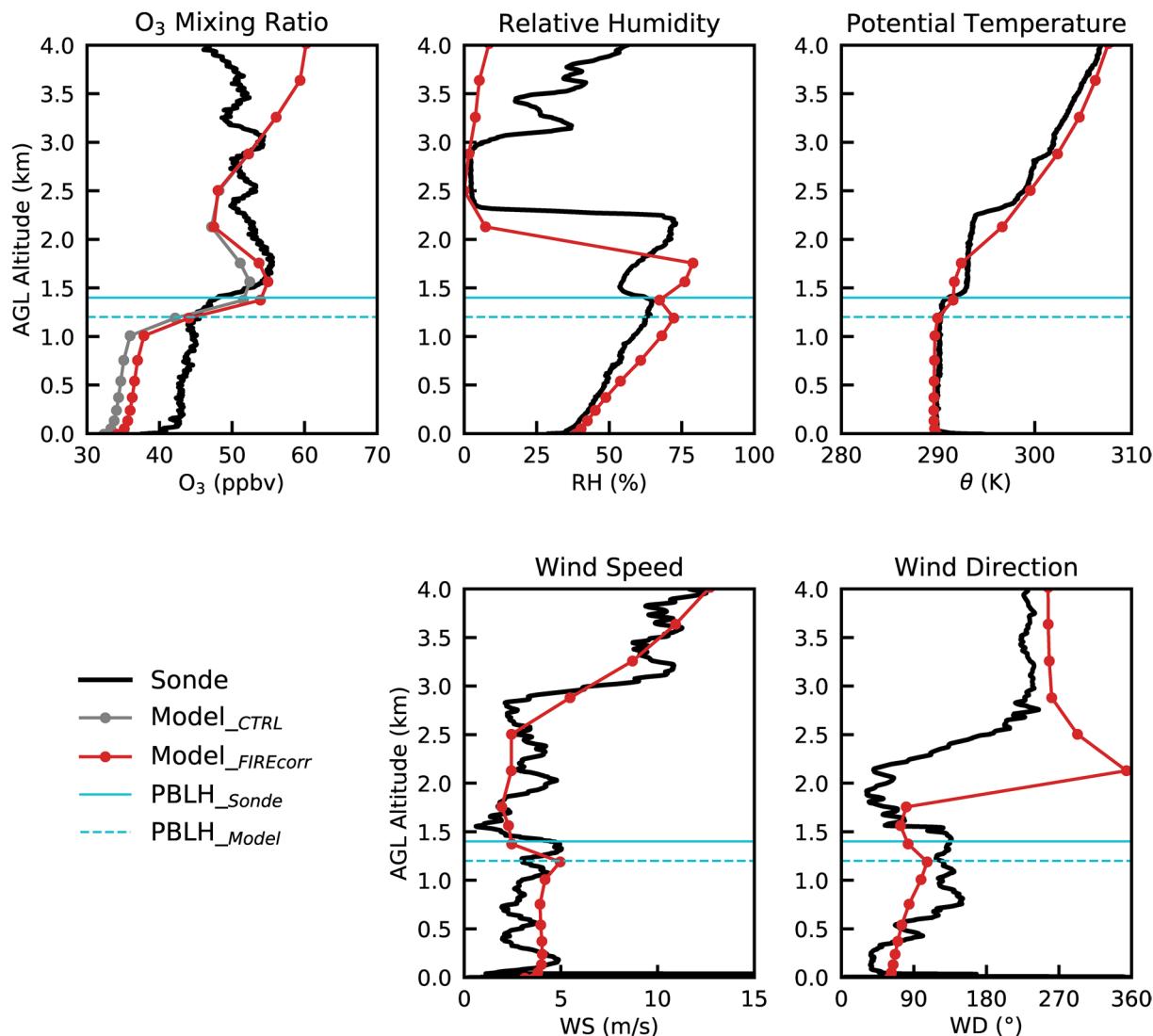
Figure 8 shows the time series of observed and modeled O<sub>3</sub> in sensitivity simulations. The model generally reproduces the observed surface O<sub>3</sub> level (below 60 ppb) during November 12–14 at most sites with a domain-wide mean bias of +4.7 ppb against hourly observations from all six sites. The results show consistent diurnal variations between simulated and observed O<sub>3</sub> at both rural and urban sites, with strong to moderate correlation coefficients (see pattern statistics in Figure S6). Among all the sites, the normalized standard deviation and correlation coefficient of domain-averaged O<sub>3</sub> are 1.0 and 0.6, respectively. As for individual sites, the model performance at Sites 3 and 5 is weaker, indicated by a low correlation coefficient and high normalized root-mean-square error. Interpreting uncertainties in modeled O<sub>3</sub> is complicated because of impacts from meteorology, emissions, and model parameterizations. The uncertainty in fire emissions is only one of many possible sources for the uncertainties in modeled O<sub>3</sub>. Other factors, such as the transport bias during the frontal passage, the model's capability in reproducing the nocturnal stable layer, and the accuracy of anthropogenic and biogenic gaseous emissions, might induce larger model uncertainties. The



**Figure 8.** Comparison between EPA (black line) and WRF-Chem ozone in November 12–14, 2016 for control run (light blue), before correction (red), and after correction (brown). Pattern statistic can be seen in Figure S6. Six EPA  $O_3$  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.

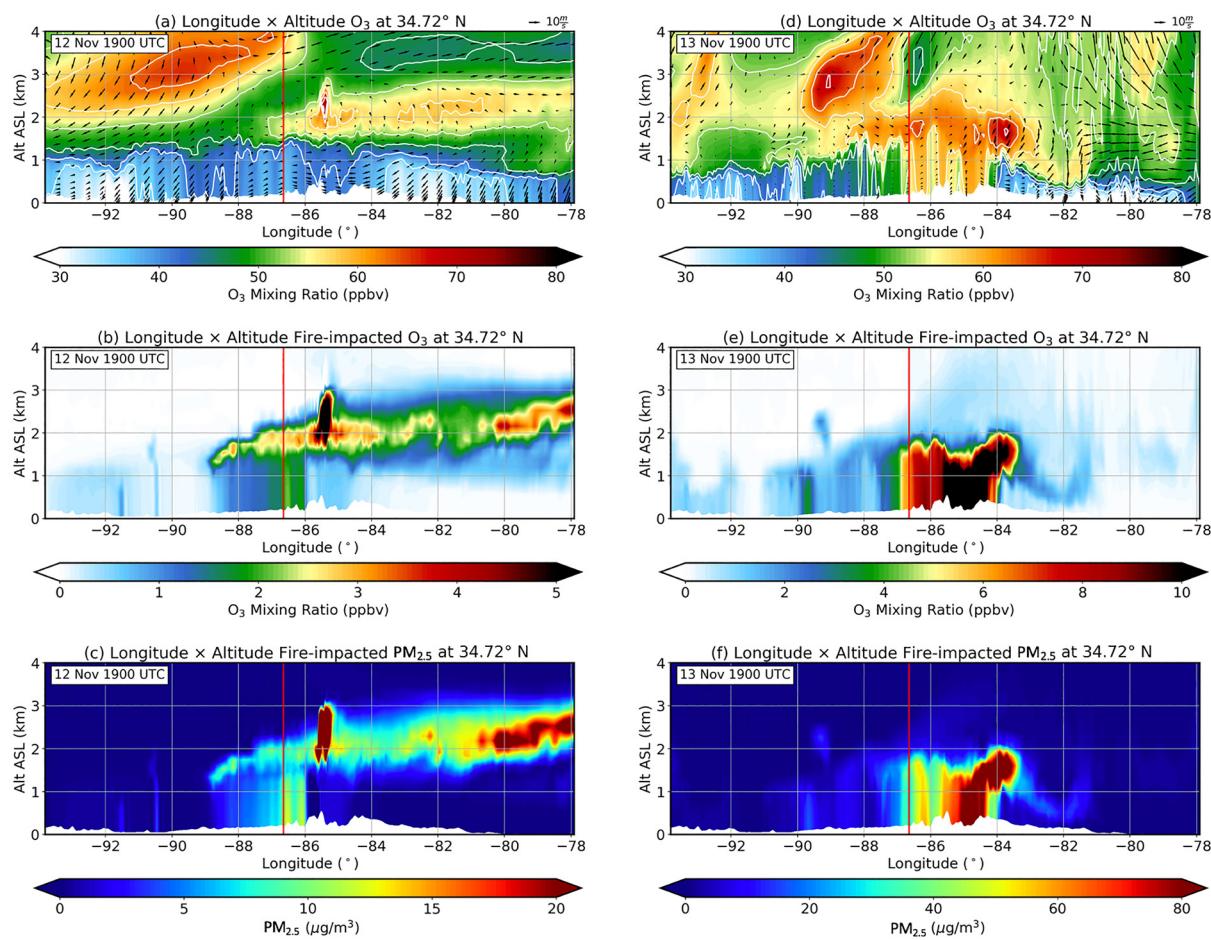
difference between FIREcorr and CTRL simulations suggests that the total fire impact on surface  $O_3$  concentration was less than 10 ppb at most sites.

An ozonesonde was launched from the UAH campus at 13:00 LT on November 12. Figure 9 compares modeled results with the observed vertical profiles, including ozone volume mixing ratio, relative humidity, potential temperature ( $\theta$ ), horizontal wind speed, and horizontal wind direction. The ozonesonde reveals an enhanced  $O_3$  lamina between two  $\theta$  inversion layers between 1.4 and 2.3 km. It peaks at 1.8 km AGL with 56 ppb, ~12 ppb larger than concentrations in the PBL. This thick lamina co-existed with the elevated aerosol plume observed in the ceilometer under a light northeasterly wind. The coexistence of a fire-impacted aerosol plume and enhanced  $O_3$  suggests that wildfires contributed to the ozone lamina above the PBL. The difference between the CTRL and FIREcorr simulations indicates a slight ozone enhancement due to fire emissions at 13 LT over Huntsville, which is further analyzed at a regional scale in Section 3.5.1. Overall, WRF-Chem is able to reproduce vertical ozone and meteorological profiles in smoke plumes below 3 km. In particular, the model reproduces the wet and ozone-rich lamina, and simulates temperature and



**Figure 9.** Comparison between ozonesonde (black color) and WRF-Chem simulations (FIREcorr in red color, CTRL in gray color) at 19:00 UTC (13:00 LT) on November 12, 2016. Ozone volume mixing ratio ( $O_3$ ), relative humidity (RH), potential temperature ( $\theta$ ), 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). Meteorological profiles and PBL height between FIREcorr and CTRL simulations are identical because the aerosol-radiation feedback is turned off.

wind field consistent with observations; however, it is limited in simulating the finer inversion layers. The model predicts a slightly lower PBL height (1.2 km compared to 1.4 km), and it does not resolve the upper  $\theta$  inversion at 2.3 km well. This limitation is likely due to the relatively coarse vertical resolution at  $\sim 2$  km and the bias in predicting wind shear when the wind turned sharply above the lamina, as observed by sonde. The underestimate in  $O_3$  in the PBL is consistent with an underestimate in surface  $O_3$  at the nearby site. This underestimate can be due partly to the model bias in wind direction and relative humidity in the PBL, as well as other factors discussed earlier. The model also reproduces the  $O_3$  laminae observed by DIAL during 19:00–23:00 LT on November 13 (Figure 7), but it underestimates the  $O_3$  magnitude in the nocturnal boundary layer, which underestimate might be caused by the uncertainties in emission inputs as discussed in the previous section.



**Figure 10.** (a) Modeled (FIREcorr) vertical sections of O<sub>3</sub> mixing ratio (ppb) along an east-west transect (the magenta line shown in Figure 1) across Huntsville latitude (34.72°N) at 19:00 UTC (13:00 LT) on November 12, 2016 at 0–4 km ASL altitude. Solid red line denotes the longitude of Huntsville. Arrows indicate modeled direction and speed of the horizontal wind. (b) Same as Figure (a), but for fire-impacted (FIREcorr minus CTRL) O<sub>3</sub> mixing ratio. Note the colorbar range is different from Figure (a). (c) Same as Figure (b), but for fire-impacted PM<sub>2.5</sub> concentration (µg/m<sup>3</sup>). (d–f) Same as Figure a–c, respectively, but for 19:00 UTC (13:00 LT) on November 13, 2016.

### 3.5. Diagnosing Fire Impacts on Vertical O<sub>3</sub> and PM<sub>2.5</sub> Distribution

Because the model performs well in simulating the vertical and surface ozone distributions and reproduces the well-mixed aerosol during the daytime, we use the model to further analyze the vertical ozone accumulation in fire smoke during the daytime on November 12 and 13, 2016. We begin with a regional sensitivity analysis to show the overall fire impacts and the possible smoke sources, and then apply the model's tendency diagnostics to examine the processes contributing to the ozone accumulation over Huntsville.

#### 3.5.1. Regional Sensitivity Analysis of Vertical Fire Impacts and Possible Smoke Sources

Figure 10 shows the modeled longitude-altitude curtain plots of O<sub>3</sub>, fire-impacted O<sub>3</sub>, and fire-impacted PM<sub>2.5</sub> at 13 LT (19:00 UTC) on November 12 and 13 over the SEUS region. On November 12, the curtain (Figure 10a) shows that an enhanced O<sub>3</sub> lamina at 2 km ASL spreads widely from 78°W to 88°W and passes over the Huntsville station. This thick layer is spreading above the PBL and is capped below 3 km by a strong wind shear when the wind turns strongly westerly above ~3 km. Figures 10b and 10c show the modeled fire impacts (FIREcorr minus CTRL) on O<sub>3</sub> and PM<sub>2.5</sub>, respectively. The simulations show enhanced O<sub>3</sub> concentrations within the elevated smoke plume, consistent with our observation analysis. Quantitatively, the fires result in an O<sub>3</sub> enhancement range of 2–5 ppb and a PM<sub>2.5</sub> enhancement range of 10–20 µg/m<sup>3</sup> at 13 LT along the west to the east cross-section. At 85°W–86°W, the enhancements estimated in O<sub>3</sub> and PM<sub>2.5</sub> can exceed 5 ppb and 20 µg/m<sup>3</sup>, respectively. Using the modeled hourly PM<sub>2.5</sub> and AOD (not shown here),

we estimate that the smoke plume is transported to Huntsville from multiple wildfires that occurred during the frontal passage on November 11 (see the large active wildfires in Figure 2 and wildfire map in Figure 1a). The wind-shear structure caps the mixed smoke plume with enhanced O<sub>3</sub> in the lowest level of FT. Below the elevated plume, there is slightly lower PBL O<sub>3</sub> enhancement (1–2 ppb) and PM<sub>2.5</sub> enhancement (5–15 µg/m<sup>3</sup>) at 86°W–88°W. The smoke in the PBL is relatively fresh with <6 h transport time and is likely emitted from nearby small fires on November 12.

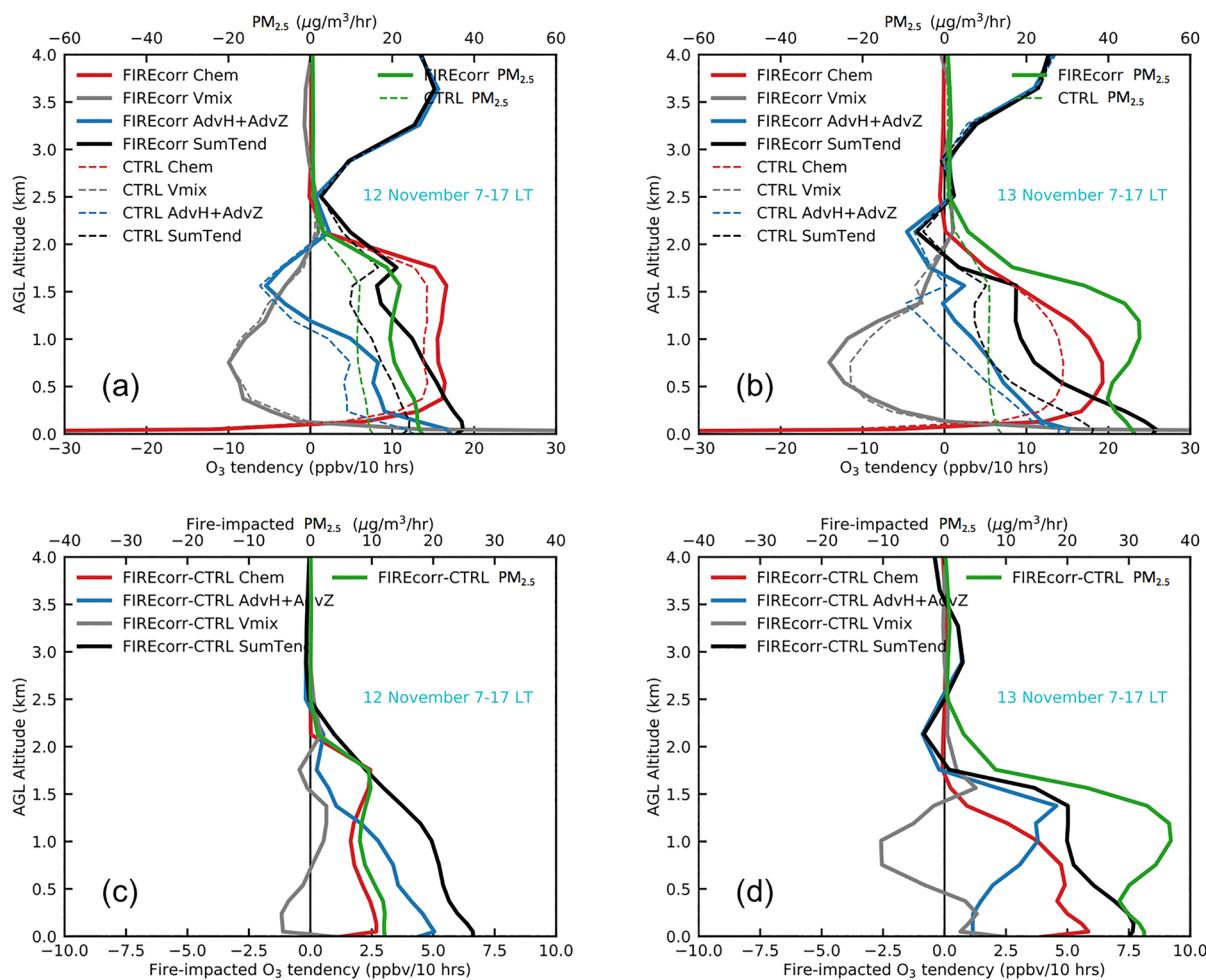
As the weather conditions turn to a high-pressure circulation on November 13, a new pattern emerges with concentrated fire impacts from the surface up to 2 km ASL on a regional scale (Figures 10d–10f). PBL O<sub>3</sub> increases in the stagnant air (Figure 10d), and the contribution of fires to O<sub>3</sub> and PM<sub>2.5</sub> increases (Figures 10e and 10f). Quantitatively, the fires result in a dominant O<sub>3</sub> enhancement range of 4–10 ppb or higher and a PM<sub>2.5</sub> enhancement range of 40–80 µg/m<sup>3</sup> or higher at 13 LT along the west to east cross-section. A large portion of the well-mixed PBL smoke is emitted on November 12, when the wildfires were most active during our study period (Figure 2). As illustrated in the observation analysis, the smoke remains in the residual layer overnight and can effectively be transported to affect other locations on the next day.

### 3.5.2. Local Process Analysis of Daytime-Integrated and Diurnal Vertical Ozone Budget in Fire Smoke

Our sensitivity simulations confirm that fire emissions impacted the vertical ozone contribution over Huntsville on November 12 and 13. This local enhancement could be caused by the transport of fire-related ozone and/or ozone precursors which increased local ozone production. The modeled results in Section 3.4 also imply that fires were not the only source contributing to the observed ozone laminae. This result brings up two questions: (1) What are the relative roles of chemical and dynamical processes on vertical ozone accumulation? (2) What is the relative contribution of fire emissions to the total net photochemical ozone production? To address these questions, we analyze the processes affecting the vertical ozone distribution using the WRF-Chem tendency diagnostics, including net chemical ozone production PO<sub>3</sub> (Chem), horizontal and vertical advection of ozone (AdvH + AdvZ), vertical mixing of ozone (Vmix), and the sum of all process tendencies (SumTend). The daytime ozone tendency output from the sensitivity simulations with fire emissions (FIREcorr) and without fire emissions (CTRL) is used to explore the fire contribution. The following model results are averaged over 5×5 horizontal grids (20 × 20 km) over Huntsville for better representativeness.

Figures 11a and 11b show daytime-integrated (07:00–17:00 LT) O<sub>3</sub> process tendencies and PM<sub>2.5</sub> concentrations over Huntsville for the FIREcorr and CTRL simulations on November 12 and 13, respectively. Here, the FIREcorr and CTRL SumTend indicate the daytime-integrated change of ozone concentration ( $\Delta O_3$ ) in simulations with and without fire emissions, respectively. The absolute O<sub>3</sub> process tendencies show similar patterns on both days. In the upper air (0.2–2.0 km AGL), the positive PO<sub>3</sub> dominates the daytime ozone accumulation on both days. The total PO<sub>3</sub> peaks by 17 ppb/10 h at 1.6 km on November 12 and by 19 ppb/10 h at 0.5 km on November 13. In the surface layer below 0.2 km, pronounced negative PO<sub>3</sub> is caused by the quick NO<sub>x</sub> titration near the surface (modeled NO<sub>x</sub> ~15 ppb). However,  $\Delta O_3$  peaks near the surface by 18 ppb/10 h on November 12 and by 25 ppb/10 h on November 13. The negative PO<sub>3</sub> near the surface is offset by positive O<sub>3</sub> contributions from vertical mixing and advection processes. Vertical mixing contributes positively near the surface yet negatively in the upper air, because it tends to disperse the enhanced O<sub>3</sub> from the upper air to the surface (Hu et al., 2019). The results imply that local chemical processes dominate the upper air ozone accumulation while dynamical processes directly contribute to the ground-level ozone build-up over Huntsville.

Figures 11c and 11d extract the relative O<sub>3</sub> tendencies and PM<sub>2.5</sub> concentrations attributable to fires (FIREcorr minus CTRL) for November 12 and 13, respectively. Here, FIREcorr-CTRL SumTend indicates the daytime-integrated  $\Delta O_3$  due to fire emissions (Figure S8). During November 12–13, fire emissions increase the vertical O<sub>3</sub> concentrations by affecting local chemical reactions, transportation, and the vertical airmass exchange. The daytime  $\Delta O_3$  due to fire emissions is similar on both days, which peaks near the surface at 7 ppb/10 h on November 12 and at 8 ppb/10 h on November 13. However, the dominant processes contributing to the total signals show daily and vertical variability. On November 12, an increase of positive PO<sub>3</sub> dominates the upper-level (above 1.2 km) fire-impacted O<sub>3</sub> accumulation, while the transport process dominates at the lower level. In contrast, on November 13, an increase in PO<sub>3</sub> (either through increased



**Figure 11.** (a and b) Process analysis of daytime-integrated vertical ozone tendencies and daytime-averaged  $\text{PM}_{2.5}$  over Huntsville in simulations with (FIREcorr, solid lines) and without (CTRL, dashed lines) fire emissions during 07:00–17:00 LT on November 12 and 13, respectively. Processes include chemical reactions (Chem, red), horizontal and vertical advectives (AdvH + AdvZ, blue), vertical mixing (Vmix, gray), and summed tendencies of all processes (SumTend, black).  $\text{PM}_{2.5}$  is represented by green lines. (c and d) are the same as Figure (a and b) but for fire-impacted values, calculated by the difference between FIREcorr and CTRL simulations.

ozone chemical production or a decrease in ozone chemical loss) dominates the lower level (below 1.0 km), while transport processes dominate at the upper level. The decrease of negative  $\text{PO}_3$  in the surface layer (i.e.,  $\text{PO}_3$  is more negative in CTRL compared to FIREcorr below 0.2 km) is affected by additional NOx and VOCs from the fires.

Quantitatively, the percentage contribution from fire emissions is calculated by (FIREcorr-CTRL)/FIREcorr during daytime over Huntsville. Fire emissions contribute 14% to the highest daytime  $\text{PO}_3$  on November 12 (2 ppb out of 17 ppb at 1.6 km) and 25% on November 13 (5 ppb out of 19 ppb at 0.5 km). This different photochemical production is associated with variable fire emissions, different smoke transport patterns, and meteorological conditions. At the surface, fire emissions contribute to the daytime  $\Delta\text{O}_3$  with 37% (7 ppb/10 h out of 18 ppb/10 h) on November 12 and with 30% (8 ppb/10 h out of 25 ppb/10 h) on November 13, respectively. In the upper air, the relative contribution increases up to 44%–58%. The smoke strength is indicated here by the fire-impacted  $\text{PM}_{2.5}$ . At the surface, fire emissions contribute to hourly  $\text{PM}_{2.5}$  with 44% ( $12 \mu\text{g}/\text{m}^3$  out of  $27 \mu\text{g}/\text{m}^3$ ) on November 12 and with 70% ( $32 \mu\text{g}/\text{m}^3$  out of  $47 \mu\text{g}/\text{m}^3$ ) on November 13. In the upper air, the relative contribution increases up to 51%–77%. The results suggest an increased fire contribution to the enhancement of ozone and particulate matter from the surface to the upper air and from one day to the next.

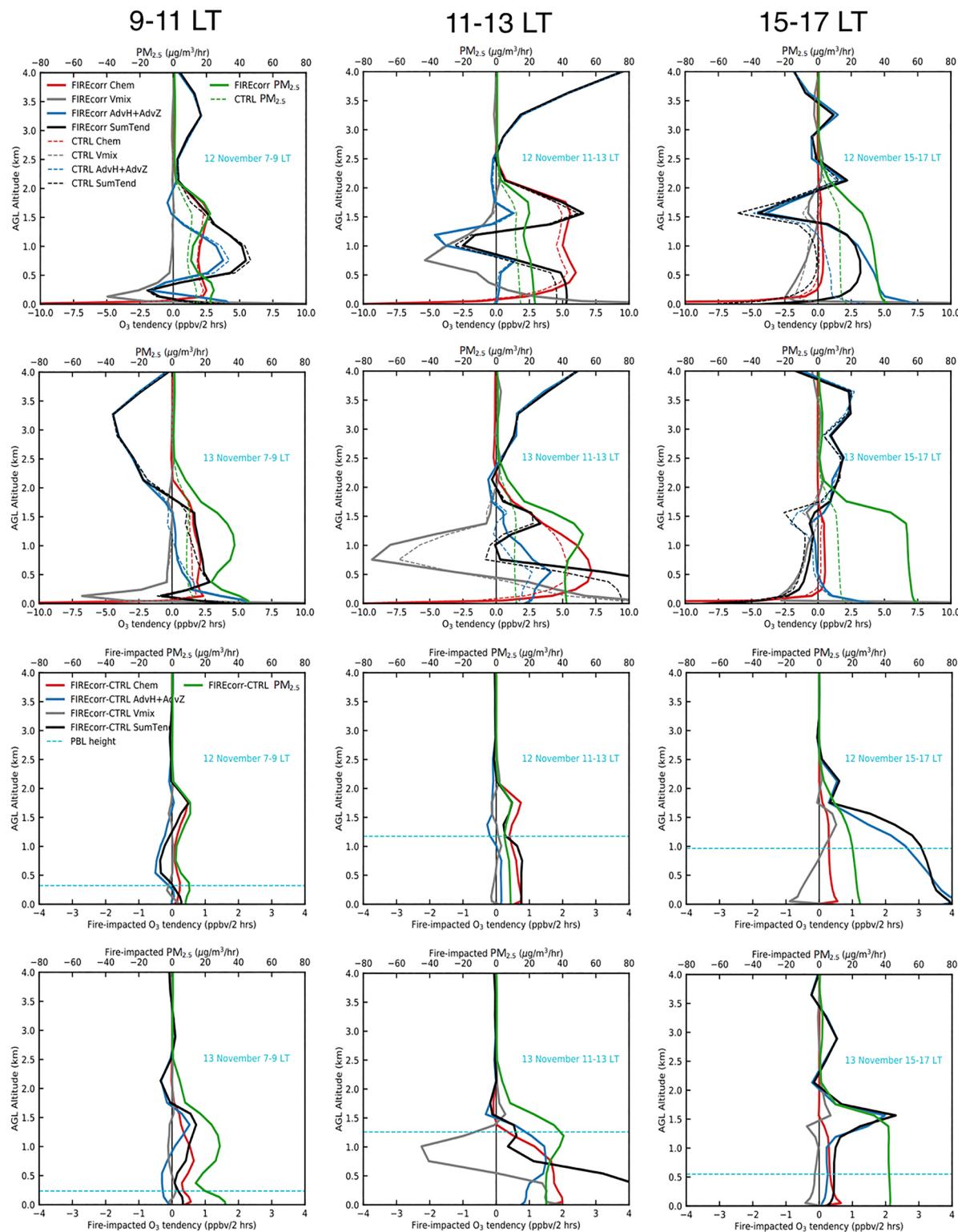
Diurnal variability of process tendencies can be affected by the boundary layer evolution, transport changes over the course of the day, and photochemistry. To examine how the different processes vary over the day, we analyze the total and fire-impacted process tendencies for 07:00–09:00, 11:00–13:00, and 15:00–17:00 LT in Figure 12. The total  $\text{PO}_3$  clearly peaks in the mid-day. The total advection term dominates in the late afternoon on November 12, and the middle of the day on November 13 when the largest inflow of ozone occurred. The total vertical mixing process is strongest when the PBL is built up in the middle of the day, and it dominates the surface ozone accumulation by dispersing upper air ozone downward.

The diurnal variability of the total tendency terms can help explain what processes drive the fire-related ozone increase during different times of the day. On November 12, the transport process in the late afternoon drives the largest fire impacts on  $\text{O}_3$  accumulation ( $\sim 4 \text{ ppb}/2 \text{ h}$  near the surface), which is associated with the freshly emitted smoke plume discussed. Fire-impacted  $\text{PO}_3$  from morning to mid-day is small, yet local net chemical production dominates the ozone increase in the elevated smoke plume. On November 13, the largest ozone increase ( $> 4 \text{ ppb}/2 \text{ h}$ ) in the mid-day is dominated by local photochemistry, followed by strong vertical mixing as well as inflow of  $\text{O}_3$  from fire emissions. This combined effect of net chemical production, transport, and vertical mixing on  $\text{O}_3$  accumulation is most pronounced in mid-day when fire smoke impacted the boundary layer.

### 3.6. Uncertainties and Limitations

In this study, turning off the aerosol-radiation feedback in WRF-Chem ensures identical meteorology between the sensitivity simulations. We acknowledge that excluding the feedback may influence modeled ozone photochemistry. To address uncertainties induced by excluding the direct radiative effect, three additional sensitivity simulations ('CTRL', 'FIREorig', and 'FIREcorr') are performed with the aerosol-radiation feedback turned on. Figure S10 shows results for photolysis rates for  $\text{NO}_2$  ( $J(\text{NO}_2)$ ) during the daytime on November 12 and 13, 2016.  $J(\text{NO}_2)$  is shown as an indicator for the rate-limiting step in the photochemical formation of ozone (Baylon et al., 2018). In addition, fire-impacted  $\text{PM}_{2.5}$  is used as an indicator for smoke altitude and magnitude. The daytime mean  $J(\text{NO}_2)$  in the three different simulations (yellow lines) shows that increasing aerosol emissions tends to slightly increase  $J(\text{NO}_2)$  at the top and above the smoke plumes, while decrease  $J(\text{NO}_2)$  in the middle to the bottom parts of the plumes. The strongest suppression of  $J(\text{NO}_2)$  appears at the bottom part of the plume. These results suggest that the additional smoke aerosols lead to a change in the vertical structure of the photochemical ozone formation, consistent with previous studies (Alvarado et al., 2015). Quantitatively, increasing fire aerosol emissions from the fires reduces surface  $J(\text{NO}_2)$  by as much as  $-4\%$  and  $-13\%$  on November 12 and 13 (red lines in Figure S10), respectively. This change in actinic flux suggests that underestimation or overestimation of photochemical ozone production largely depends on the altitude with respect to smoke plumes for a given fire event. Our sensitivity simulation suggests that, by excluding the aerosol-radiation feedback, daytime ozone at the surface might be overestimated by  $4\%$  and  $13\%$  on November 12 and 13, respectively. A slight overestimation in the plume center and a slight underestimation above the plume may result from excluding the feedback. Jiang et al. (2012) examined the direct radiative effects on ozone production for large-scale fires. Their results show that the reduction in both downward shortwave radiation and surface temperature can decrease both  $J(\text{NO}_2)$  as well as biogenic isoprene emissions, resulting in reductions in surface ozone concentrations by as much as  $15\%$ .

Our analysis has limitations due to excluding SOA and heterogeneous/aqueous chemistry in the MOZCART scheme. The formation of SOA in smoke plumes is highly variable; SOA can increase as the plume ages while in other cases can stay constant or even decrease (Akagi et al., 2012; Alvarado et al., 2015; Wigder et al., 2013). In a recent model study for the 2016 SEUS wildfires, Guan et al. (2020) have shown that SOA from fires accounts for  $9\%$  and  $12\%$  of  $\text{PM}_{2.5}$  components for November 6–9 and November 13–16, respectively. Therefore, considering SOA, our adjustment factor of aerosol emissions might be reduced from 3.6 to  $\sim 3.2$ . Future studies are needed to estimate the additional uncertainties to the simulated AOD but are not expected to significantly alter our conclusions, namely that a factor of 3–4 is needed to scale the FIN-Nv1.5 aerosol emissions for this fire event. Heterogeneous chemistry in aerosols and clouds may also affect the ozone budget in different ways. Although there is little evidence for significant heterogeneous ozone loss, the uptake of  $\text{HO}_2$  and  $\text{N}_2\text{O}_5$  by aqueous aerosols represents a potential sink (Jacob, 2000). A comparison between the most recently added T1 MOZCART and the MOZCART scheme used here shows that



**Figure 12.** Same as Figure 11, but present diurnal variability by integrating 2 h in 7–9, 11–13, and 15–17 LT on November 12 (Rows 1 and 3) and November 13 (Rows 2 and 4), respectively.

T1 MOZCART generally simulates somewhat lower surface ozone than MOZCART (<https://www2.acom.ucar.edu/wrf-chem>), and this change is mostly attributed to the inclusion of heterogeneous reactions on aerosols in T1 (Emmons et al., 2020). Although the results may differ when simulating other regions or times, we do not expect that the omission of heterogeneous chemistry will significantly alter our results. In addition, ceilometer measurement and satellite images show that the sky over Huntsville during our study period is mostly clear except for a short-time cloudy condition on the morning of November 13. Therefore, we expect a small impact on local ozone results from cloud chemistry for this specific case study.

#### 4. Conclusions

Analyzing measurements from ozonesonde, O<sub>3</sub> DIAL, ceilometer, surface monitors, and space-borne observations together with the regional chemical transport model WRF-Chem provides evidence of wildfire impacts on the vertical distribution of ozone and aerosols over Huntsville, AL. Diagnostic analysis characterizes the relative roles of chemical and dynamical processes in the vertical ozone budget in fire smoke. Multi-platform observations are shown to be essential to evaluate the model performance from regional to local scale. This study uniquely combines the TOLNet/RO<sub>3</sub>QET UV O<sub>3</sub> DIAL aerosol extinction product with MODIS AOD and EPA PM<sub>2.5</sub> concentrations to compare with the modeled smoke during a high particulate pollution episode of the 2016 SEUS wildfires.

During the daytime on November 12 and 13, 2016, fire emissions contribute 12–32 µg/m<sup>3</sup> (40%–70%) to hourly surface PM<sub>2.5</sub> at Huntsville and dominate the local particulate air pollution on November 13. Besides the freshly emitted smoke plumes, relatively aged plumes emitted from previous days contribute considerably to the entire PBL PM<sub>2.5</sub> accumulation. Fire emissions contribute 7–8 ppb/10 h (30%–37%) to the daytime surface ozone increase at Huntsville. Although O<sub>3</sub> is of less concern for air quality during this fire episode because concentrations remained below national health standards, investigating the fire influence mechanism on the vertical ozone budget provides insights into the variability of the ozone distribution downstream of wildfires. In particular, the main sources of fire-impacted ΔO<sub>3</sub> are demonstrated by sensitivity simulations and tendency diagnostics: (1) Fire emissions increase the vertical ozone concentrations downstream of the fires by affecting the local net chemical ozone production, inflow, and outflow of ozone, and the vertical ozone exchange. These processes vary in importance on daily, diurnal, and vertical scales. On November 12, local net photochemical ozone production over Huntsville dominates the fire-impacted ozone enhancement in the elevated plume while transport processes dominate the boundary layer ozone accumulation in the late afternoon. On November 13, local net photochemical ozone production dominates the fire-impacted ozone enhancement below 1 km. (2) Local net chemical production of ozone from biomass burning was not the dominant source of surface ozone these days. However, the vertical mixing and advection of ozone produced elsewhere from biomass burning emissions enhanced the overall impacts of biomass burning on local ozone. The combined effect of chemical and dynamical processes leads to a percentage contribution to ΔO<sub>3</sub> of 30%–37% at the surface and up to 44%–58% in the upper air.

For the considered case studies, WRF-Chem captures the general day-to-day AOD pattern, air-quality variations, vertical structure of aged plumes, and enhanced ozone lamina. Three main avenues for future work follow from our work: (1) Discrepancies in fire emission estimations need to be considered for model inputs. The (OC + BC)/CO emission ratios in FINNv1.5, GFEDv4s, GFASv1.2, and QFEDv2.5\_r1 fire inventories differ by a factor of 5.7 (in Gg per Gg) over the 2016 SEUS wildfire region. A scaling ratio of 3–4 on aerosol emissions (derived from FINNv1.5 PM<sub>2.5</sub> and PM<sub>10</sub>), within the spanned range of the emission ratios in different inventories, was needed in our case to improve the modeled magnitude of surface PM<sub>2.5</sub>, vertical aerosol extinction, and AOD. (2) After the emission adjustment, the underestimation of the densest plume in DIAL and highest AOD in MODIS is partly due to missing fire detections under clouds on November 13. Adding extra satellite detections (e.g., FINNv2.2 includes VIIRS information) or filling in the gap of missing fire counts in emission estimation algorithms should be considered; (3) The density, continuity, and species of vertical measurements are relatively limited for modeling evaluation. Available larger samples of vertical measurements (ground-based and airborne) with ambient data will benefit regional-model evaluation in future fire studies.

In summary, our results reveal the different mechanisms by which fires can influence the vertical ozone budget downstream of the wildfires and point out large uncertainties in fire emissions. Our case study reveals the benefits of combining observations from multiple platforms for characterizing fire impacts on surface and upper air chemical composition and for the in-depth evaluation of models. Albeit ozone concentrations during the considered fire episode were of less concern for air quality management, our analysis of the ozone budget in fire smoke provides valuable insight into the complex interplay of chemical and dynamical processes. Future work combining modeling diagnostic tools with a larger sample of ground-based and airborne measurements for a multitude of fire episodes is needed to gain a more generalized understanding of the ozone evolution in wildfire plumes and subsequent air quality and policy implications.

## Data Availability Statement

The ceilometer datasets used in this study are archived at <https://www.nsstc.uah.edu/mips/data/data/>. The ozonesonde and DIAL datasets in this study are available at <https://doi.org/10.5281/zenodo.4019663>. Surface ozone and PM<sub>2.5</sub> datasets are downloaded from the Environmental Protection Agency (EPA) website (<https://www.epa.gov/outdoor-air-quality-data>). MODIS aerosol optical depth data is acquired from the NASA Earth Data Level-1 and Atmospheric Archive & Distribution System Distributed Active Archive Center (LAADS DAAC) website via <https://ladsweb.modaps.eosdis.nasa.gov/>. 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>.

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