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# JGR Atmospheres

## RESEARCH ARTICLE

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### Key Points:

- Stratospheric intrusions can contribute 10 to 25 ppbv to the background ozone
- Biomass burning plumes can contribute 10 to 80 ppbv to the background ozone
- Biomass burning in the western United States can significantly increase Midwest pollution levels

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(continued)

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## Evaluation of Stratospheric Intrusions and Biomass Burning Plumes on the Vertical Distribution of Tropospheric Ozone Over the Midwestern United States

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**Abstract** Naturally occurring ozone-rich stratosphere-to-troposphere transport (STT) intrusions and biomass burning (BB) plumes reaching the surface can contribute to exceedances of the U.S. National Ambient Air Quality Standards for ground-level ozone (70 ppbv implemented in 2015). Additionally, fires can inject significant pollution into the free troposphere where it can be transported long distances. The combined air quality impacts from these sources on ozone have only been analyzed in a few case studies for the Midwest United States. Here we study ozone impacts in a Midwestern city, for the first time in St. Louis, Missouri, using a series of ozonesonde profiles taken during the Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC<sup>4</sup>RS) field campaign in August–September 2013. All ozonesondes showed enhancements above the background profile levels (~55 ppbv) throughout each tropospheric column. Two models were used to estimate ozone origins in columns. A chemical transport model identified STT enhancements equivalent to 10 to 15 ppbv over the background with a 10% to 15% contribution overall to the column. Two FLEXPART-WRF simulations, one with smoke in the boundary layer and another with smoke above, identified BB enhancements equivalent to 10 to 80 ppbv. Overall, the total BB contribution is 15% to 30% of the total column. Five ozonesondes showed signatures of mixed BB plumes and STT intrusions. During this study period, BB in the western United States contributed 70% to ozone enhancements in the total column compared to 3% from the central United States and 27% from other areas.

**Plain Language Summary** Because man-made emissions are decreasing, concentrations of harmful ozone pollution have also decreased in many areas of the United States. Not all sources can be easily controlled however. For example, biomass burning emits lots of pollutants to the atmosphere, and descending air from the stratosphere can bring with it high levels of ozone. These sources can pollute first the air above us, and then when the air is transported to the surface, it can pollute the air we breathe. In this study we used balloons to measure ozone pollution as it changes from the surface to high in the atmosphere. We then used computer models to identify the sources responsible for higher pollution levels. Our study was part of a major field campaign that took place in the Midwest United States in the summer of 2013.

## 1. Introduction

Even moderate concentrations of ground-level ozone (O<sub>3</sub>) can adversely affect human health and the environment across the United States (U.S. Environmental Protection Agency [U.S. EPA], 2015). To lessen these impacts, the U.S. EPA (2015) has lowered the National Ambient Air Quality Standards (NAAQS) for O<sub>3</sub> from 75 to 70 parts-per-billion-per-volume (ppbv). Ozone is a key secondary air pollutant associated with a large number of health issues ranging from asthma to premature deaths (Fann et al., 2013, 2018; Liu et al., 2015, 2017; Rappold et al., 2017; Reid et al., 2016), leading to 12,300 to 52,000 annual premature deaths in North America alone (Silva et al., 2013). Due to more stringent regulations over the past 20 years, regional ozone levels have declined by 7% to 13% (Simon et al., 2015;

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Strode et al., 2015). New evidence has demonstrated that even at lower concentrations ozone can still be very toxic (U.S. EPA, 2013). Despite overall progress, ozone concentrations may still be increasing in some areas (Cooper et al., 2015; Fishman et al., 2014; Jaffe et al., 2018), particularly in fire-prone regions (McClure & Jaffe, 2018).

Regulating locally formed ozone is complicated by the fact that ozone has significant background levels in the troposphere typically ranging 30 to 50 ppbv (Jaffe et al., 2018). Background concentrations of ozone can be enhanced due to noncontrollable ozone sources (NCOS), defined by Jaffe et al. (2018) to include recent local pollution influence from stratosphere-to-troposphere transport (STT), lingering biomass burning emissions, long-range transport from international sources, lightning, or photochemical production from precursor emissions (e.g., nitrogen oxides and volatile organic compounds). Therefore, in this study we define background ozone to include NCOS. While foreign sources of pollution and wildfires are theoretically controllable, these are beyond the control of any local jurisdiction, so for this discussion, we can justify including these as background ozone. Over the past decade, significant progress has been made in our efforts to understand aspects of the U.S. background ozone problem, for example, episodic stratospheric sources (Lin et al., 2015), interannual variability (Strode et al., 2015), and wildfire contributions (Jaffe et al., 2013; Westerling, 2006, 2016). However, these efforts have lacked coordination and are largely focused on the impacts to the western United States where NCOS are considered to be the greatest (Jaffe et al., 2018; Jaffe & Wigder, 2012). Despite a 15% to 33% reduction in ozone precursors from anthropogenic emissions across the Eastern and Midwestern United States (Strode et al., 2015), the Midwestern background ozone levels continue to increase  $\sim 0.23$  ppbv per year (Fishman et al., 2014) on par with the trends in the intermountain western United States.

The Midwestern United States, like the intermountain west United States, is frequently impacted by biomass burning (Liu, Mickley, et al., 2016; McCarty et al., 2007) and stratospheric intrusions (Langford et al., 2009, 2018; Lin et al., 2015). Although biomass burning in the Midwest consists mostly of smaller agricultural fields, their combined emitted pollution and higher frequency can result in emissions double that of wildfires (Larkin et al., 2014). Understanding stratospheric intrusions and biomass burning contributions for this region will be critical as they can confound NAAQS attainment (e.g., Hess & Zbinden, 2013; Jaffe, 2011; Lin et al., 2015), where summertime daily ozone maxima already range from 70 to 80 ppbv and occasionally reach mixing ratios higher than 140 ppbv (Fishman et al., 2014). The periodic nature of some NCOS makes it difficult to target these sources with dedicated field campaigns, but opportunistic measurements have been made during field studies with other objectives (Ott et al., 2016; Sullivan et al., 2015). For example, the influence of wildfires, long-range transport, and STT were the foci of the Las Vegas Ozone Study (LVOS) where it was demonstrated that NCOS contributed  $\sim 30$  ppbv to three ozone exceedances (Langford et al., 2015).

The Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC<sup>4</sup>RS) field campaign in August–September 2013 presents a unique opportunity to study Midwest U.S. background ozone (Toon et al., 2016). During the flight mission several ozonesondes were launched nearly every day to measure vertical profiles of column ozone and meteorology over seven U.S. stations in a network known as SouthEast American Consortium for Intensive Ozonesonde Network Study (SEACIONS). The SEAC<sup>4</sup>RS campaign sparked a growing number of studies concerning ozone in the southeast United States (Liu, Zhang, et al., 2016; Travis et al., 2016; Wagner et al., 2015; Zhu et al., 2016). Although the research flights did not cover the Midwest United States, this area is of interest because the time of the field campaign was an active period for NCOS with signatures for STT, large fires in the western United States, and smaller prescribed fires in the central United States ( $\sim 0.2$  million hectares burned). During the 2013 ozone season (1 April through 31 October), 15 total ozone exceedances occurred with six high ozone days occurring during SEAC<sup>4</sup>RS. Additionally, a satellite-based detection method identified smoke plumes with confirmed injection heights above the boundary layer, initiated by smoke-infused thunderstorms known as pyrocumulonimbus or pyroCb (Fromm et al., 2010, 2019; Peterson et al., 2015, 2018; Peterson, Fromm, et al., 2017; Peterson, Hyer, et al., 2017). The parametrization of plume rise from biomass burning can vary widely and cause undue biases and errors within model simulations of pollution transport (Paugam et al., 2016; Val Martin et al., 2012, 2018; Wilkins et al., 2018). The detection algorithm used for the smoke plumes reduced the uncertainty of the injection heights and gave improved estimates based on remote sensing of each specific case.

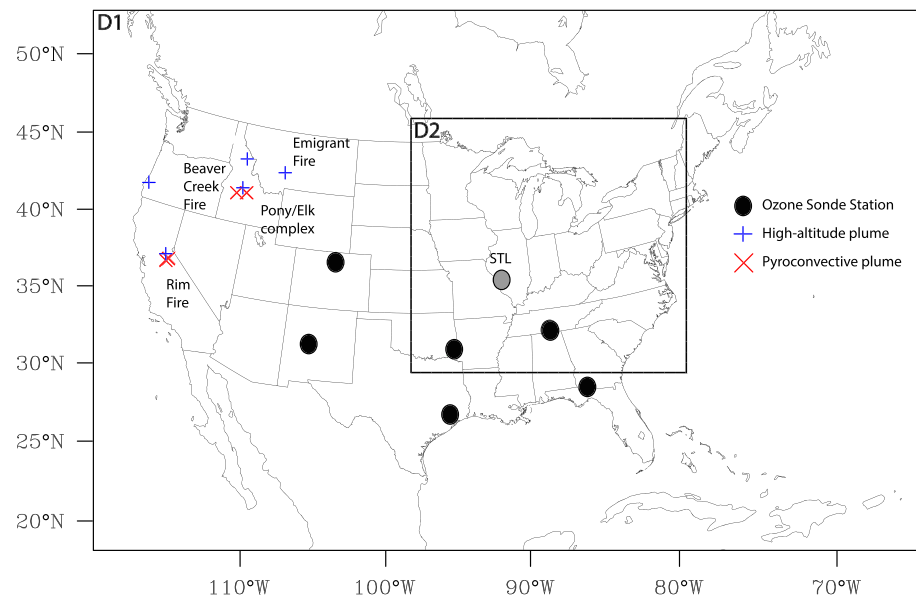
Using a chemical transport model, it has been shown that fires can contribute ~14% to daily max ozone levels in the central United States (Wilkins et al., 2018). This has driven the need for developing accurate tropospheric ozone budgets for a typical summertime in the Midwest as suggested by Wilkins et al. (2018). Several approaches have been used to partition or identify the individual contribution of a naturally occurring ozone-rich STT intrusions (Langford et al., 2018), a biomass burning plume (Westerling, 2016), or their combined impacts (Brioude et al., 2007) to elevated ozone levels. These approaches include the use of intensive balloon-borne ozonesonde field campaigns (Cooper et al., 2007; Thompson et al., 2019); lidar and aircraft remote sensing (Brioude et al., 2007; Langford et al., 2018); and Lagrangian trajectory and chemical transport models (Baker et al., 2016, 2018; Morris et al., 2006). However, the determination of the contribution from individual sources to local ozone enhancement with a single point measurement remains difficult (Thompson et al., 2019), and it can be even more challenging to quantify accurately the contribution from multiple sources (Lin et al., 2015). In particular, the partitioning of these sources is critical for NAAQS attainment investigations along with long-term trend determinations, as they are two of the most critical NCOS imported (Parrish et al., 2012, 2014). Further, 30% of the present-day atmospheric ozone burden is attributable to human activity, and these emissions are still rising (Granier et al., 2011; Parrish et al., 2010). For example, Li et al. (2019) using ~1,000 surface ozone sites over a 5-year period (2013–2017) showed that East Asian concentrations are continuing at +1 to 3 ppbv per year. In this study, we use ozonesonde measurements from the SEAC<sup>4</sup>RS campaign and models to identify and characterize contributions from biomass burning and stratospheric intrusions to tropospheric ozone columns above the Midwest background ozone levels. Additionally, we evaluate the contribution from fires observed in the western and Midwest United States to ozone concentration in St. Louis.

## 2. Tropospheric Ozone and Meteorological Soundings

Vertical profiles of ozone, pressure, humidity, and temperature were measured using balloon-borne ozonesondes coupled with radiosondes. Each ozonesonde consists of a Teflon pump, an ozone sensing electrochemical concentration cell (ECC) (Komhyr et al., 1986, 1995), attached alongside a Vaisala RS-8015N radiosonde. The instruments were prepared in accordance with the Southern Hemisphere Additional Ozonesondes (SHADOZ) protocol (Thompson, Allen, et al., 2011; Thompson et al., 2019). The ECC-type ozonesonde is currently the most widely used due to its well-characterized behavior and working capability under various sky conditions (Kuang et al., 2012; Thompson, Oltmans, et al., 2011). Comparisons with other O<sub>3</sub> measuring instruments have demonstrated the ECC sonde precision to be  $\pm 5\%$  near the ground and  $\pm 10\%$  in the upper troposphere, although errors can reach 17% (Stauffer et al., 2014; Thompson et al., 2007). The radiosonde temperature and pressure sensors have accuracies below 20 km of  $\pm 0.3^\circ\text{C}$  and  $\pm 0.5$  hPa, respectively (Lal et al., 2014). The heights were calculated based on the observed pressure and are above sea level (asl). The humidity sensor has an accuracy of about  $\pm 2\%$  near the ground which decreases to  $\pm 15$ –30% in the 5- to 15-km altitude range (Hurst et al., 2011; Kley et al., 1996).

Figure 1 displays the seven ozonesonde sites across the United States for the SEACIONS — archived online (at <http://croc.gsfc.nasa.gov/seacions/>). Here, we focus on the sole Midwestern United States site located at the James S. McDonnell Planetarium in Forest Park (90.27°W, 38.63°N, 181 m asl), ~5 km west of downtown St. Louis, Missouri. Balloon soundings carrying an ozonesonde and a radiosonde were made near daily between 10:00 and 14:00 (UTC – 5 hr). The exact launch time depended on clearance from the Air Traffic Control of the local airport and weather conditions. The first flight was made on 8 August 2013 and the last on 22 September 2013. There were breaks scheduled between the launches in order to cover the time period of the larger SEAC<sup>4</sup>RS flight mission (Toon et al., 2016). A total of 28 ozonesondes was launched during this period from St. Louis. The balloons reached a maximum altitude of 22 to 36 km with an average ascent rate of about  $\sim 5$  ms<sup>-1</sup>. The instruments fell mostly within 70 km of the launch site.

Vertical curtain plots of ozone concentrations are shown in Figure 2, displayed in 500-m bins up to 15-km height. The figure shows structures in the tropospheric ozone profile which will be analyzed for impacts of stratospheric air masses and biomass burning sources using the modeling techniques demonstrated by Thompson et al. (2007) and Thompson, Oltmans, et al. (2011).

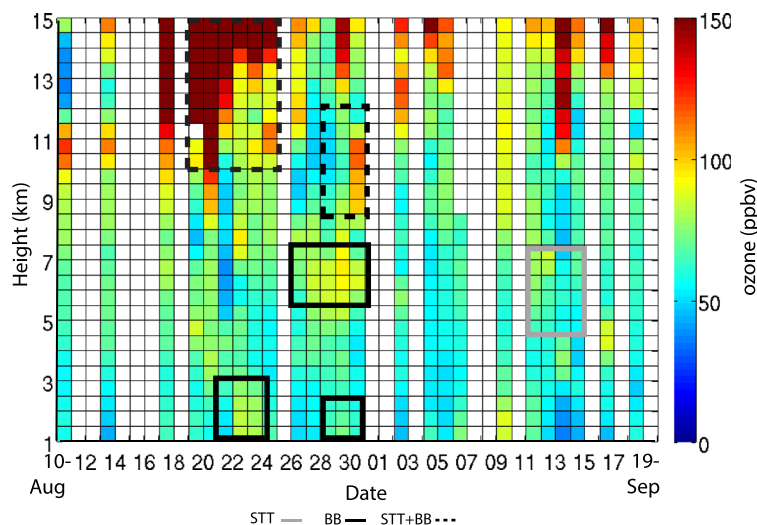


**Figure 1.** Study area map with FLEXPART-WRF model domain. Rectangles mark the WRF domains D1 which has a horizontal resolution of 12 km and D2 (4-km resolution). SEACIONS locations are marked by black circles (St. Louis, Missouri station [STL] is a gray circle). Wildfire emissions from FLAMBE, August–September 2013, with 11 pyroconvective and 5 high-altitude injection elevated smoke plume activity areas are marked; red crosses indicate pyroCb plumes which transport smoke into the upper troposphere and lower stratosphere. Blue crosses indicate high-altitude injection of smoke in the absence of large pyroCb which transport smoke into the middle troposphere.

### 3. Numerical Models

#### 3.1. Stratospheric-to-Tropospheric Transport Ozone Tracer Model

To simulate the stratospheric air masses entering the troposphere during the SEACIONS field campaign, the NASA Goddard Earth Observing System model (GEOS-5, Rienecker et al., 2008) was used. The model



**Figure 2.** Ozonesondes in support of SEACIONS were launched at the Saint Louis University ozonesonde station located at the James S. McDonnell Planetarium in Forest Park (90.27°W, 38.63°N, 181 m asl), 5 km west of downtown St. Louis. Vertical tropospheric profiles at ~18:30 UTC (1:30 p.m. local time) ozone profiles are averaged vertically every 500 m and shown below the thermal tropopause level (~15 km). Cases of ozone enhancements above the background (~55 ppbv) are shown by source origin: the gray box for stratospheric-to-tropospheric transport (STT), the solid black line boxes for biomass burning (BB), and the dashed line black boxes for combined STT and BB.



analysis was provided on 72 levels from the surface to 0.01 hPa (~75 km), every 6 hr. Initial conditions for the atmospheric component were taken from uncoupled experiments forced by the observed sea surface temperature. The model defines a stratospheric intrusion as air masses with a high ozone composition, potential vorticity greater than 1.5 potential vorticity units (PVU;  $1 \text{ PVU} = 10^{-6} \text{ m}^2 \text{ s}^{-1} \text{ K kg}^{-1}$ ), and a relative humidity (RH) of less than 20% (e.g., Holton et al., 1995; Stohl, 2003; Waugh & Funatsu, 2003). Here we categorize each STT by temporal duration, vertical extent, and meteorological cause and relate them to the contributions to the tropospheric ozone column. Previously, GEOS-5 has been used extensively for stratospheric intrusions and has been validated with ozonesonde observations (e.g., Stajner et al., 2008; Wargan et al., 2015). A review of data assimilation methodology applied to chemical constituents, including ozone, can be found in Lahoz et al. (2007).

### 3.2. Dispersion Model

#### 3.2.1. FLEXPART-WRF Lagrangian Dispersion Model

The FLEXPART-WRF version 3.1 (Brioude et al., 2013) was used as a Lagrangian particle dispersion model using wind fields from the Weather Research and Forecasting (WRF version 3.4, Skamarock et al., 2008). Simulations were made for 7 days to relate ozonesonde profiles to biomass burning events. FLEXPART-WRF was used to simulate carbon monoxide (CO) as a passive tracer for biomass burning plumes. The plume injection heights were adjusted to match the impacts with the vertical ozone profiles at a resolution of 100 m. The model does not include chemical loss or production in its trajectory calculations. The simulations can be used to estimate CO concentrations using multiple particles each representing a fixed amount of CO emitted and transported as a passive scalar. Therefore, trajectories aged 5 to 15 days can be considered as mixing ratios above background (Brioude et al., 2007). WRF was initialized using the North American Regional Reanalysis (NARR, Mesinger et al., 2006) wind fields, with hourly temporal resolution, two nested domains with a horizontal resolution of 27 and 9 km, and 40 pressure levels. Figure 1 shows a map of the two nested WRF domains. Nine individual WRF simulations were performed to cover the entire time span of the SEACIONS mission. Each simulation lasted 162 hr with 42 hr of spin-up time, and the remaining 5 days were used for the analysis (de Foy et al., 2014).

To determine the transport impacting each ozone measurement, a plume origin path was calculated. The plume path was determined based on several thousand forward trajectory particles released from a box surrounding the location and time of biomass burning present over the 44-day period. Plume origin path distributions were output in 1-day intervals on a  $2.5^\circ \times 2.5^\circ$  output grid at 500-m vertical resolution covering the United States, with a  $0.25^\circ \times 0.25^\circ$  at 100-m vertical resolution nested grid over St. Louis. FLEXPART-WRF outputs the plume origin paths in units of  $\text{s kg}^{-1} \text{ m}^3$ , which represents the residence time of the plume per grid cell divided by the air density. The residence of each plume was calculated for the surface to upper troposphere (~0 to 15 km) layers of the atmosphere. This is known as a weighted plume residence time (Lal et al., 2014).

The dispersion of a plume origin path forward in time indicates the likely source regions contribution of the ozone precursors to the measured ozone but over the previous 7 days. For plumes passing through these layers, the residence time of the forward trajectories can interact with stratospheric intrusions allowing for the detection of mixed plumes. Hourly biomass burning emissions were provided by the Fire Locating and Modeling of Burning Emissions (FLAMBE) program (<http://www.nrlmry.navy.mil/flambe/>) (Reid et al., 2009). The emission factors are outlined by Ferek et al. (1998). FLAMBE provides quasi-operational, with fire location, instantaneous estimates of fire size, and smoke emission flux in  $\text{kg m}^{-2}$  generated in near real time for the Western Hemisphere (Reid et al., 2009). The inventory uses active fire detection from the GOES's Wildfire Automated Biomass Burning Algorithm (WF-ABBA) and MODIS's Active Fire Products to detect biomass burning activity in near real time (Reid et al., 2009). The fire emissions are calculated in grams of CO from the inventories' burned area ( $\text{m}^2$ ) and smoke aerosol emissions (CO in  $\text{g m}^{-2}$ ) variables. The fire emissions are mapped onto a  $0.25^\circ \times 0.25^\circ$  grid as input to FLEXPART-WRF. With this technique, the quantity of CO emitted into each plume from several fire source regions across the United States was tabulated. The CO tracer has no chemical or depositional removal processes and was treated as a passive tracer. The NARR analysis contains information that can be used to derive stratospheric ozone values above and within the troposphere. We used these values to calculate the quantity of ozone transported from the stratosphere to the location where a plume was released. NARR provides more information on the

**Table 1**

*Elevated Smoke Plumes Escaping the Planetary Boundary Layer Identified by the Naval Research Laboratory Satellite Detection Algorithm for High-Altitude Injection During the Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC<sup>4</sup>RS) in the Summer 2013*

Date	Time (UTC)	Lat	Lon	State	Fire name	Plume type
7 August 2013	22:15	42.71	−123.66	OR	Big Windy Complex	High altitude
8 August 2013	N/A	43.46	−114.55	ID	Beaver Creek	Pyroconvective
8 August 2013	N/A	43.42	−114.85	ID	McCan	Pyroconvective
9 August 2013	22:41	43.29	−115.55	ID	Pony/Elk	Pyroconvective
10 August 2013	21:00	43.36	−115.45	ID	Pony/Elk	Pyroconvective
12 August 2013	21:00	43.29	−115.55	ID	Pony/Elk	Pyroconvective
12 August 2013	23:00	43.62	−114.51	ID	Beaver Creek	Pyroconvective
13 August 2013	23:41	43.50	−115.43	ID	Pony/Elk	Pyroconvective
16 August 2013	21:41	45.66	−114.78	ID	Gold Pan Complex	Pyroconvective
16 August 2013	23:41	43.62	−114.51	ID	Pony/Elk	High altitude
16 August 2013	23:41	43.46	−114.55	ID	Beaver Creek	Pyroconvective
19 August 2013	22:50	45.20	−110.69	MT	Emigrant	High altitude
19 August 2013	23:15	37.86	−120.09	CA	Rim	Pyroconvective
21 August 2013	22:41	37.86	−120.09	CA	Rim	Pyroconvective
22 August 2013	21:30	41.04	−123.47	CA	Corral Complex	High altitude
25 August 2013	17:00	37.91	−120.00	CA	Rim	High altitude

*Note.* Smoke plumes detected are categorized: (1) a pyroconvective plume containing smoke reaching the stratosphere (~10 to 15 km) or (2) a high-altitude plume containing smoke reaching into the middle troposphere (~5 to 9 km).

stratospheric intrusions structure with temporal coverage eight times daily and a 32-km spatial grid. Stratospheric intrusions in NARR were calculated using PVU distribution on isentropic surfaces, 320 to 410 K ( $\approx 80$  hPa). Specifically, with NARR we can closely match ozonesonde launch times with meteorological data (~18:00 UTC); this allows for better comparison with model and observed stratospheric intrusions.

### 3.2.2. FLEXPART-WRF High-Altitude Smoke Injection

To account for the vertical extent (or plume injection height) of varying biomass burning emissions in the simulations, we employ the FLEXPART-WRF model. The BASELINE simulation represents biomass burning emissions as a uniform release within the well-mixed boundary layer up to ~3.5 km (e.g., Brioude et al., 2007; Colarco, 2004; Davison, 2004; Lioussse et al., 1996). To represent plumes that penetrate well above the boundary layer, we use the U.S. Naval Research Laboratory (NRL) satellite-based pyroCb detection algorithm and inventory (Peterson, Fromm, et al., 2017; Peterson, Hyer, et al., 2017)—referred to hereafter as the PYRO simulation. For a confirmed pyroCb, the plume tops are set at the model determined upper troposphere and lower stratosphere (UTLS) boundary (~9 to 15 km). For non-pyroCb, plume tops are set to be near the model determined middle troposphere (~5 km). In both cases, the plume bottoms are set above the PBL, with the emissions uniformly distributed within the tropospheric model layers. Lastly, a method is presented that is a combination of the two methods mentioned above (BASELINE + PYRO) referred to hereafter as the combined method. The combined method was run as a single simulation and compared to each individual method. The combined simulation included all the emissions from the baseline, with addition of the elevated emissions from the fires with pyroconvective plumes identified in Figure 1; see Table 1 for more details.

Peterson, Fromm, et al. (2017) examined a variety of wildfires and pyroconvective events in western North America during the summer (June to August) of 2013. At least 26 intense pyroCb events were inventoried, injecting smoke particles well into the UTLS. Several fires produced smaller pyroCbs, and others injected smoke above the boundary layer in the absence of significant pyroCb activity (Figure 1). The NRL pyroCb detection algorithm is designed to detect large plumes that impact the UTLS and, in general, is not designed to capture lower tropospheric transport. The algorithm only detects anvil cloud tops; therefore, neither the exact quantitative plume height nor a plume bottom can be detected.

A variety of additional observations and methods exist to track the evolution of pyroCb smoke plumes in the UTLS after the initial injection (e.g., Fromm et al., 2010, 2019; Peterson et al., 2018). However, application of

these methods is beyond the scope of this study. Evolution and transport of significant pyroCb plumes from the 2013 fire season are examined in greater detail by Fromm et al. (2019), Peterson et al. (2015), Peterson, Fromm, et al. (2017), and Peterson, Hyer, et al. (2017).

## 4. Results and Discussions

### 4.1. The Structure of Tropospheric Ozone Columns

The vertically averaged near-daily profiles of ozone are shown in Figure 2. The profiles are averaged every 500 m vertically from the surface to the tropopause (~15 km) with annotations for enhanced ozone-rich plumes, above the tropospheric column mean (55.7 ppbv), from biomass burning and stratospheric intrusions. Ozone gradually increases with altitude in the lower troposphere (below about 3.5 km). Average ozone is about 55 ppbv near the surface but decreases to 45 ppbv near 3.5 km. Above 3.5 km, ozone increases slowly throughout the entire troposphere at a rate of 3 ppbv km<sup>-1</sup>. In the middle troposphere (4- to 10-km height), average ozone is highest in August and lowest in September. While above 10 km, ozone is similar for both months. August profiles averaged 10 to 15 ppbv higher than September. Ozone is lowest (~40 ppbv) during the month of September near the surface and remains low below 3.5 km. However, on 5 September the second highest recorded surface ozone day occurred at the surface (60-ppbv ozonesonde, 95 ppbv nearby surface monitor later in the day). The maximum measured ozone from a sonde at the surface occurred 30 August at 65 ppbv. Other notable enhancements (NAAQS exceedances) of O<sub>3</sub> (>70 ppb near the surface) occurred on 23 August, and 4–6 and 10–11 September. A mean PBL of 2.4 km and a mean thermal tropopause at 14.9 km could be clearly identified in the ozonesonde profiles. Note that these figures for biomass burning contributions from boreal fire contributions to ozone downwind are somewhat larger than the 5% to 15% estimates of this quantity by Thompson, Allen, et al. (2011; see Figure 13) or Moeini et al. (2020) that are based on Canadian soundings. These last two studies attribute 20% to 25% of the ozone column amounts to stratospheric origins.

### 4.2. Stratospheric-Tropospheric Transport Tracer Impacts

Table 2 lists the GEOS-5 determined stratospheric intrusions events that impacted St. Louis during August and September 2013, expressed as deep or shallow with the pressure level of 600 hPa being the threshold. The 44-day study period consisted of 13 events. The intrusions recorded resulted from either a shortwave trough (6 days: 7, 10, 23, 28, and 30 August and 6 September), frontal passage (7 days: 3 and 12 August and 3, 12, 15, and 22 September), or cutoff low (1 day: 14 August). Three frontal pass intrusions reached the lower troposphere and penetrated the boundary layer. About 40% of the intrusions reached the middle troposphere (~500 hPa). Stratospheric intrusion generally lasted 1 to 3 days occurring either early morning before 3:00 (UTC – 5 hr) or around noon local time. The intrusions were found to contribute on average 10 to 25 ppbv to tropospheric ozone columns which equates to around 5% to 15% of the total tropospheric ozone column.

### 4.3. Biomass Burning Emissions

Figure 3 shows the hourly U.S. total mass of fire smoke emissions from FLAMBE in Gg-CO km<sup>-2</sup> hr<sup>-1</sup> gridded per 0.25° cell with a minimum threshold of 2,000 kg. From 8 August to 22 September 2013 the calculated total emissions were 1,060 Gg of CO. FLAMBE emissions depict a regional distribution pattern in known fire regions: agricultural and prescribed fires in the southeast United States (e.g., the Carolinas, Florida, and Georgia) and southcentral United States (e.g., Louisiana, Mississippi, and Arkansas) and the wildfires in the central plains and intermountain west United States (e.g., Wyoming, Idaho/Montana area, and California). A time series of the hourly emissions indicates that August contained almost all the major fire emissions (Figure 4). The duration of the largest fire emissions is indicated in the figure by arrows. Between 14 and 24 August 2013, on average ~6 Gg-CO hr<sup>-1</sup> (~1.2 × 10<sup>4</sup> FLEXPART particles where 1 particle = 500 kg = 0.5 × 10<sup>-3</sup> Gg) was being emitted daily with a maximum hourly total of 11 Gg-CO hr<sup>-1</sup> (~2.2 × 10<sup>4</sup> FLEXPART particles). Regional fires emissions from the central United States were considerably lower than wildfires emissions in the intermountain west (3% compared to 70%).

Specifically, three large wildfires areas were identified that produced the most emissions and high-altitude smoke injection plumes. The Idaho Beaver Creek fire, caused by a lightning strike on 7 August 2013 in



**Table 2**  
*Simulated Stratospheric Air Masses Entering the Troposphere (Stratospheric Intrusion) Over St. Louis, Missouri, During August and September 2013*

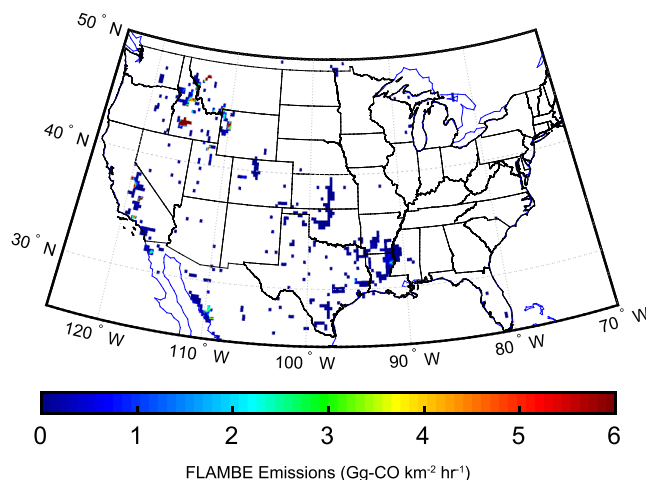
Date	Time (UTC)	Duration (hr)	Height (hPa)	Extent	Meteorology
3 August 2013	03:30	21	580	Shallow	Frontal passage
7 August 2013	13:30	28	690	Deep	Shortwave
10 August 2013	02:30	34	800	Deep	Shortwave
12 August 2013	18:30	28	500 to 850	Deep	Frontal passage
14 August 2013	06:30	180	600	Shallow	Cutoff low
23 August 2013	16:30	9	500	Shallow	Shortwave
28 August 2013	21:30	18	480	Shallow	Shortwave
30 August 2013	08:30	94	500	Shallow	Shortwave
3 September 2013	12:30	30	700 to 1000	Deep	Frontal passage
6 September 2013	09:30	41	600	Shallow	Shortwave
12 September 2013	16:30	10	800	Deep	Frontal passage
15 September 2013	20:30	48	800 to 1000	Deep	Frontal passage
22 September 2013	00:30	94	900	Deep	Frontal passage

the Sawtooth National Forest, occurred in the lower southwestern edge of the Elk/Pony Complex (43.59°N, 114.68°W). This fire burned an area of 465 km<sup>2</sup> (114,900 acres), emitting 54.6 Gg-CO of smoke emissions, and accounted for 14% of the Idaho and Montana states combined total fire emissions (392 Gg-CO km<sup>-2</sup> hr<sup>-1</sup>) during the months of August and September. The second fire area, led by a fire named the Montana Emigrant Fire, was a part of a small system of fires caused by multiple lightning strikes that spanned across Yellowstone National Park (45.20°N, 110.69°W) consuming <81 km<sup>2</sup> (20,000 acres) starting 14 August emitting 9.4 Gg-CO of smoke emissions. Lastly, the largest of the three, the California Rim Fire, described in detail in Peterson et al. (2015), caused by an illegal campfire, occurred on 17 August 2013 in Yosemite National Park (37.85°N, 120.08°W). The Rim Fire burned an area of 1,041 km<sup>2</sup> (257,314 acres), emitted 224 Gg-CO of smoke, and accounted for over 66% of the California biomass burning emissions (341 Gg-CO km<sup>-2</sup> hr<sup>-1</sup>) during SEAC<sup>4</sup>RS.

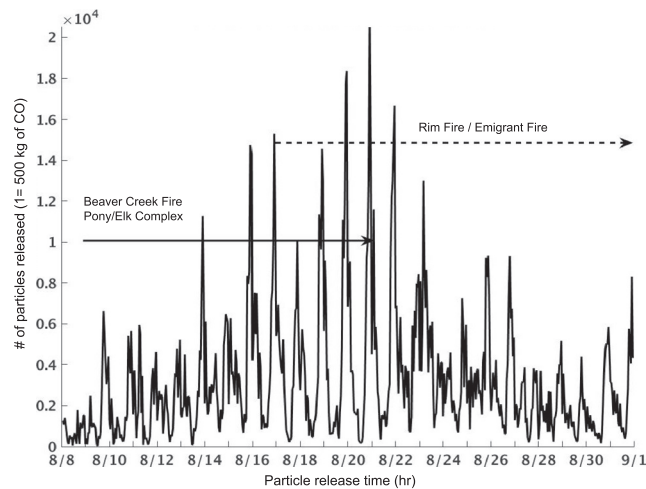
#### 4.4. Residence Time of Particles Over St. Louis

The FLEXPART-WRF plume origin path (forward trajectories) has been used to calculate residence times of air parcels above St. Louis (Figure 5). Figures 5a–5f show the average residence times of the biomass

burning plume origin paths expressed as plume age and CO tracer particle amounts as concentrations. Results are displayed for the BASELINE simulation (Figures 5a and 5d), PYRO simulation (Figures 5b and 5e), and combined (Figures 5c and 5f). The lighter shading represents a higher concentration of CO particle counts (Figures 5a–5c), and for plume age the lighter shading indicates older air (Figures 5d–5f). The major contributions below 3 km are from the central U.S. fires. The contributions from the intermountain west U.S. fires dominate above 3 km, in the range of 80% to 90%. Other regions contribute in the range of 5% to 10%. Biomass burning plumes are indicated to have impacted the surface ozone each day but with varying magnitudes. The surface impacts were connected to mechanisms causing air parcels to move downward (e.g., shortwave on 30 August). Likewise, strong vertical motion was evident in bringing simulated air within the boundary to the middle and upper portions of the troposphere. For example, on 24–26 August at the 8- to 12-km layer, both simulations indicate enhancements of 0.74 ppm of CO that has been aged ~7 days. This enhancement corresponded to ~40 ppb of excess O<sub>3</sub> at the 11- to 12-km layer in the ozonesondes. Ozone impacts for August from biomass burning sharply increase after 3 km and vary daily



**Figure 3.** Biomass burning emissions of carbon monoxide (CO) estimated by FLAMBE in units of Gg km<sup>-2</sup> hr<sup>-1</sup> in North America during SEACIONS, 8 August to 22 September 2013. FLAMBE emissions are displayed on the model grid which has a resolution of 0.25° per cell. Only cells with emissions above 2,000 kg are shown.



**Figure 4.** A time series of FLAMBE emissions (mass/time) converted to particles released in FLEXPART-WRF (# of particles or particle count). Each 500 kg of mass is converted to 1 particle in FLEXPART-WRF simulations. Locations emitting less than 500 kg only emit a single particle. Arrows represent the duration of two of the largest wildfires (Rim Fire and Beaver Creek) with relevance to SEACIONS with other fires (Emigrant Fire and Pony/Elk Complex) contributing to those time frame emissions indicated. The dotted line is the California Rim Fire (37.85°N, 120.083°W) 17–31 August 2013, and the solid line is the Idaho Beaver Creek (43.593°N, 114.684°W) 7–21 August 2013. Note: Beaver Creek and Pony/Elk Complex occurred nearly at the same time and were fewer than 100 km apart in distance. While, the Emigrant Fire contributed a significant amount of emissions during its occurrence at the same time as the Rim Fire but was much smaller in magnitude.

meteorological situations in the central United States: (i) cyclonic flow or a system of low pressure (e.g., cutoff low 17–21 August) and (ii) anticyclonic flow or a system of high pressure (e.g., blocking high 25–30 August). Table 3 lists each episode's ozone enriched plume, for two selected days (21 and 30 August), identified by an ozonesonde over St. Louis (Figure 2). For each identified ozone enhancement, the simulated source-contribution relationships from biomass burning (Table 1) and stratospheric intrusions (Table 2) are quantified. The corresponding travel time of the plume (or plume age) and concentrations are indicated by FLEXPART-WRF (Figure 5). Each plume was individually characterized with model simulations, and ozonesonde observational data in addition to the meteorological conditions leading to the plume reaching St. Louis are discussed below.

#### 4.5.1. 17–21 August 2013: Evidence of Biomass Burning Advected by a Cutoff Low

The meteorological map provided in Figures 6a–6c indicates biomass burning transported by a cutoff low originating on 17 August 2013 00:00 UTC at 500 hPa over central Missouri. In this case the cutoff low was created by a closed upper-level low pressure system that was cut off from a westerly current (14 August 06:30 UTC), and it began moving independently. The cutoff low remained nearly stationary until 19 August 00:00 UTC, at which time the trough filled in and moved eastward. Due to the nature of the quasi-stationary system, the westerly flow of air propagated the western fire plumes over the central United States and southeast United States. Figures 7a and 7c present a cross section and plane view of the resulting low. Due to the low pressure system the trajectory analysis indicated that smoke flowed from various fire locations to the St. Louis area (see Figure 8). Figures 9a–9c depict the vertical ozone and meteorological profiles representative of this time period (21 August). Refer to Figure 2 for corresponding ozonesonde curtain plots.

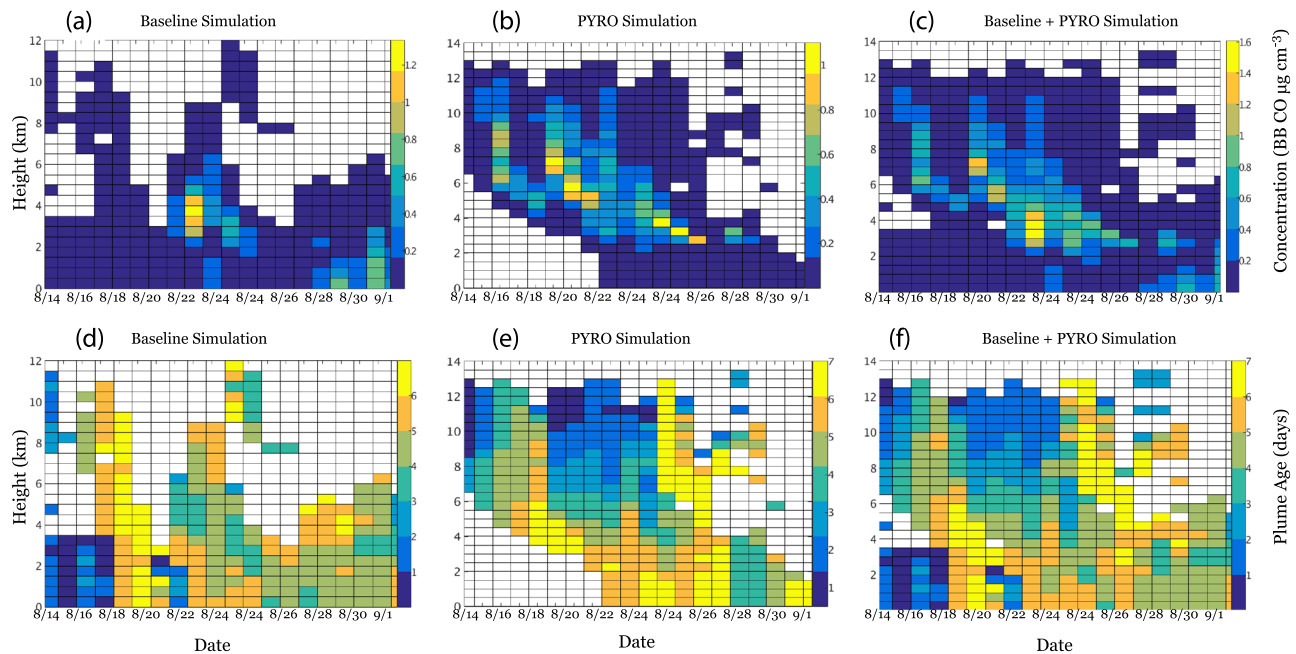
Three ozone enhanced layers are visible on 21 August (Table 3 and Figure 9). None of these enhancements are deemed of stratospheric origins, as there was no significant PVU present in these layers. Plume P3 at 11 km was initially considered to be a stratospheric intrusion related but is ruled out as the NARR data

above, up to 12 km where the biomass burning contributions to ozone decrease to near negligible amounts. The high-altitude smoke injection simulation indicates that plumes that get injected above the 4-km height can impact the surface. Evidence of aged lofted air, ranging from 4 to 15 km, impacting the surface is seen 22 August to 1 September 2013, where a constant ~2 ppm of CO enhancement at the surface is present. Additional evidence of biomass burning emissions enhancing ozone within the lower tropopause and PBL (0 to 6 km) is present on 21–24 August over the St. Louis area—a greater than 15-ppbv enhancement of ozone within a 6-day aged plume containing an additional 9 ppm of CO.

The average plume age for the August month ranged between 4 and 7 days. Plumes reaching the lower troposphere (below 3.5 km) were ~3 days fresher than plumes above. The simulated high-altitude smoke injection is typically ~2 days fresher than the boundary layer injected smoke when above 3 km but can be 2 days older when below. The average aged plumes lead to 15-ppbv increase in ozone. As plumes aged, they became more ozone enriched. Fresher plumes have higher levels of CO but lower amounts of ozone. An example of this occurred 23 August, where ozone exceedance at the surface was indicated in both simulations to be from a polluted air mass layer with mixed ages. From the lower and middle troposphere (below 1.5 km and above 5.5 km), the plumes ages averaged 2 days older, with about 7 ppm fewer CO and 20 ppbv more ozone than the air in the layers in between.

#### 4.5. High-Altitude Smoke Injection Analysis

The impacts of high-altitude smoke injection are demonstrated using two examples from commonly encountered summertime synoptic



**Figure 5.** FLEXPART-WRF biomass burning (BB) CO concentrations and age are simulated, binning trajectories over St. Louis within a  $2.5^{\circ} \times 2.5^{\circ}$  grid box and averaged vertically 500 m with daily temporal resolution. FLAMBE emissions of BB CO are converted into particle amounts and are released. (a) The base simulation, boundary layer simulation, releases emissions within the PBL, (b) NRL's adjusted pyroconversion scheme is implemented, PyroCb simulation; particles are released from pyroCbs and high-altitude injection as specified in Table 1, and (c) the combined (a) and (b). With panels (d)–(f), corresponding to plume ages in simulations. The average BB CO  $\text{g cm}^{-3}$  per grid cell (top) lighter shading indicates higher concentration, and transported CO plume ages (bottom) lighter shading indicates older air.

and sonde thermal tropopause indicate that the tropopause was lowered (15 to ~9 km) possibly by wave breaking from the cutoff low. The lower- and upper-middle tropospheric ozone enhancements at P1 (2 to 5.5 km) and P2 (7 to 9 km), respectively, are determined to be of biomass burning origin. The lower-tropospheric plume P1 was determined to be from the Idaho Beaver Creek plume ~6 days earlier. The upper-middle tropospheric plume P2 originated 2 days prior from high-altitude injection plumes from the Yellowstone National Park Emigrant cluster fires. The contribution of 21 August tropospheric ozone layers from biomass burning was 10 to 25 ppbv. The pyroCb from the Idaho Pony Elk Fire (16 August at 23:41 UTC) reached the St. Louis area. But the polluted air remained in the lower stratospheric air and did not mix with the lower layers. The incorporation of the high-altitude injection plumes allowed for the two middle- and upper-tropospheric polluted plumes to be characterized which contributed ~15% to 30% to layers in the total ozone column. This upper-level pollution was later advected down to the surface and contributed to the exceedance on the 23 August.

#### 4.5.2. 25–30 August 2013: Evidence of Stratospheric Air Mixing With Aged Biomass Burning During Anticyclonic Flow

The meteorological map provided in Figures 6d–6f indicates aged biomass burning transported into a stratospheric intrusion by a high pressure system that became well established over Missouri 25 August 00:00 UTC. The high pressure system began developing a week prior over the southcentral United States. The high remained quasi-permanent as it sat over Missouri reaching its maximum strength on 26 August 06:00 UTC until a middle-level (500 hPa) shortwave trough moved into the area on 29 August 12:00 UTC moving the system eastward. The near surface reflection of the shortwave reaches Eastern North Dakota 29 August 18:00 UTC as the trough extends south to Oklahoma where it penetrates deeper into the ridge. Figures 7b and 7d present a cross section and plane view of the resulting high. Due to the high pressure system the trajectory analysis indicated that smoke flowed from various fire locations in addition to stratospheric intrusion impacts to the St. Louis area (see Figure 8). Figures 9d–9f depict the vertical ozone and meteorological profiles representative of this time period (30 August). Further evidence of this trough effects on elevated ozone aloft in the sonde measurements (see Figure 2) remained until 30



**Table 3**

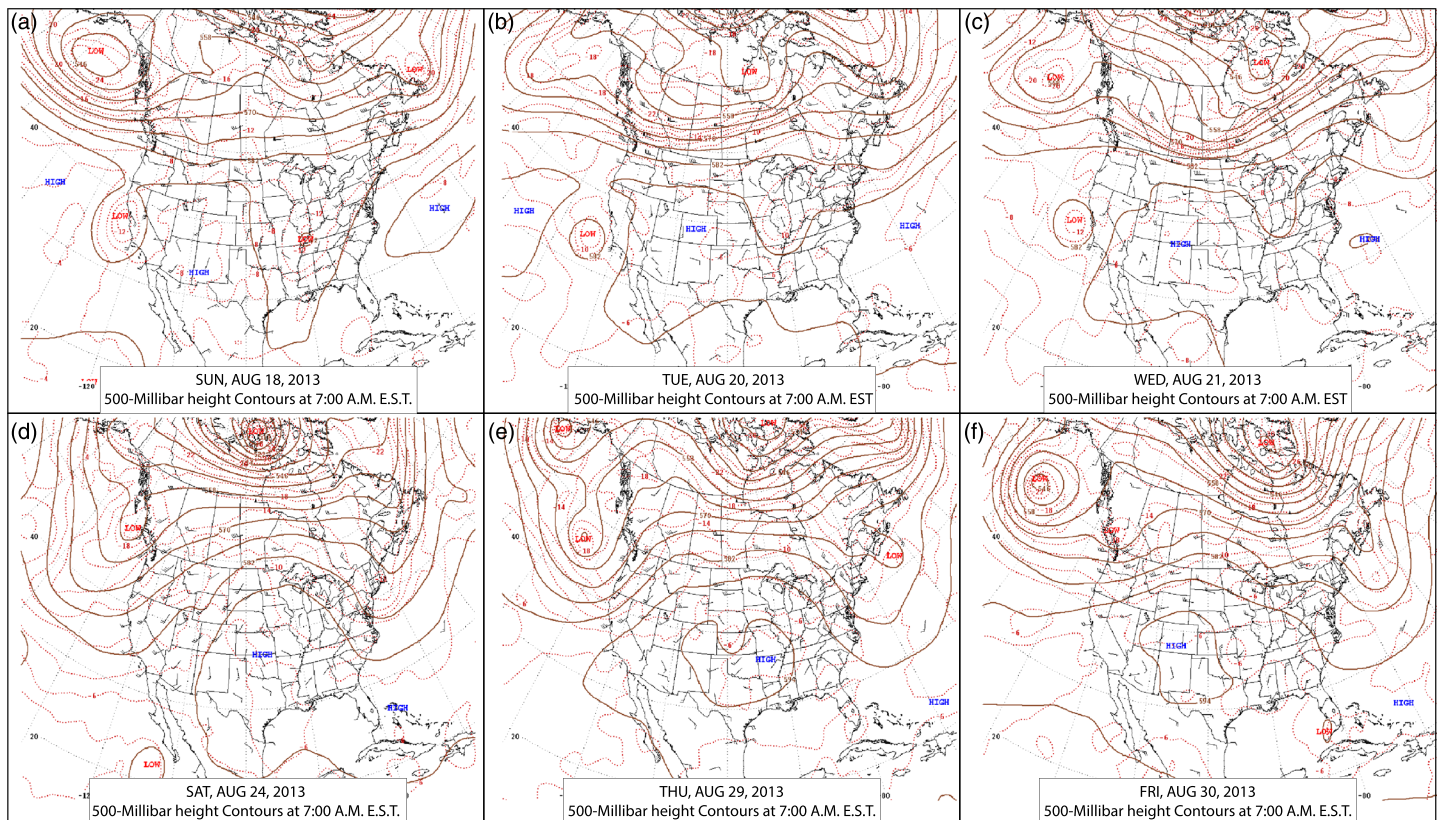
*Ozone ( $O_3$ ) Enriched Plumes Identified Over St. Louis, Missouri, Using Ozonesondes Vertical Profile Measurements for Two Episodes*

Plume ID	Date	Source	Plume height (AGL km)	Smoke age (days)	BASELINE simulation (CO ppm)	PYRO simulation (CO ppm)	Sonde RH (%)	Sonde $O_3$ (ppb)	Excess $O_3$ (ppb)
P1	21 August	ID Beaver Creek	2 to 5.5	5.8	2.4	14	10	65	10
P2	21 August	MT Emigrant	7 to 9	2.6	0	5.2	5	85	25
P3	21 August	pyroCb and SI	11	1.2	0	0.63	20	65	10
P4	30 August	SE US Ag. Fire	0 to 2.5	5.1	6.6	0.83	85	65	10
P5	30 August	Unidentified	5 to 7	6.7	0	0.35	10	100	45
P6	30 August	CA Rim Fire and SI	8 to 12	5.3	0	6.8	15	140	80

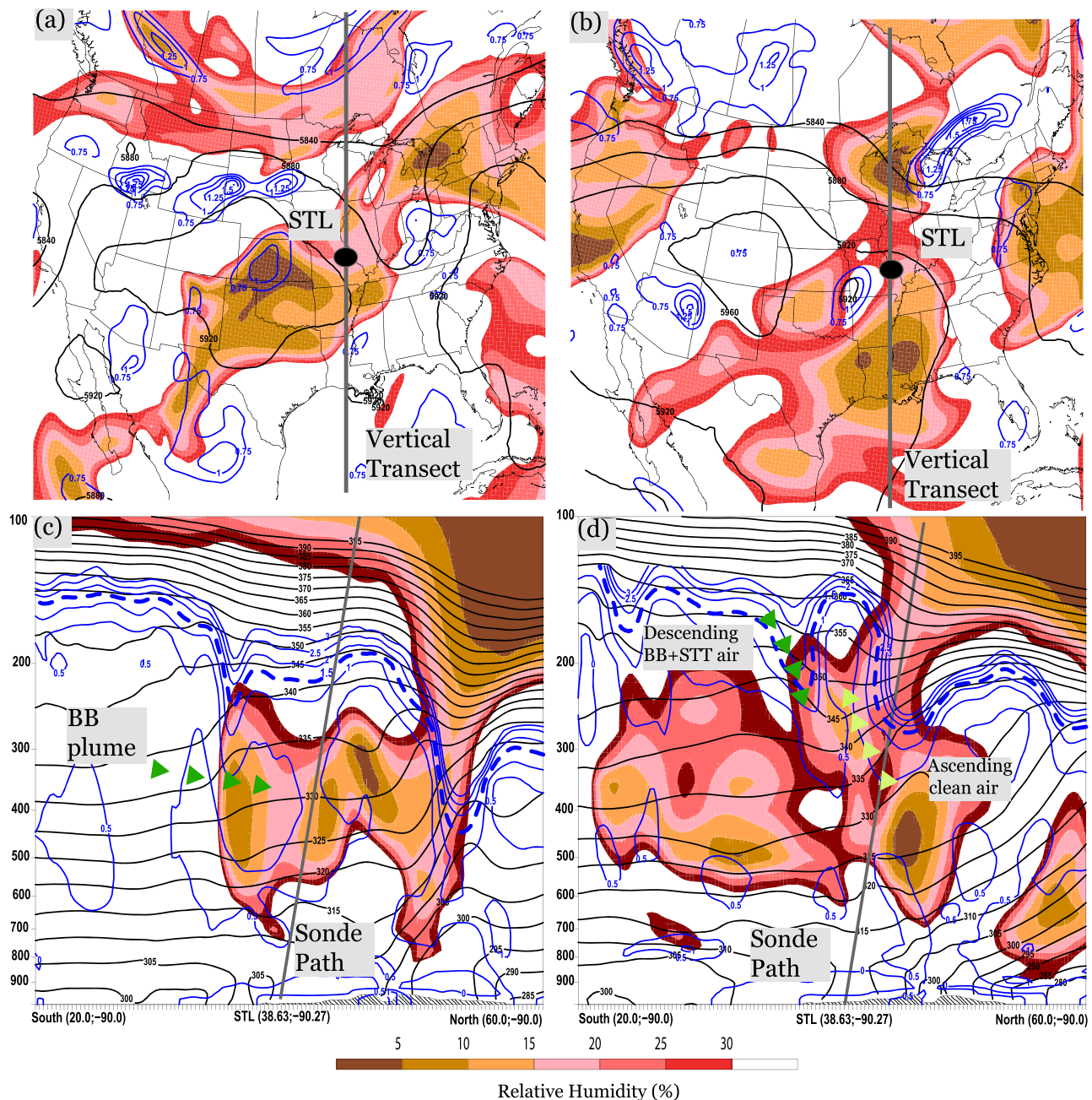
*Note.* Included are the simulated biomass burning plumes and stratospheric air masses entering the troposphere (stratospheric intrusion [SI]) concentrations of a carbon monoxide (CO) tracer for each plume. Plume ID corresponds to labeling in Figure 9.

August 18:00 UTC (Figures 9d–9f). Thereafter, a shortwave passes the area slightly to the east and cleared out the excess ozone.

Three ozone enhanced layers are present during the high pressure event as evident on 30 August (Table 3, P4 to P6). Stratospheric air descended into the middle troposphere ~500 hPa on two occasions during this time period (28 and 30 August) from shortwaves (Figure 7d). Stratospheric impacts are indicated at ~7 to 9 km and >12 km of layers with high PVU values and low RH values (Figures 9d–9f, P5 and P6). NARR PVU analysis provided further proof that these are in fact stratospheric in origin. Figure 7d depicts lowering dry



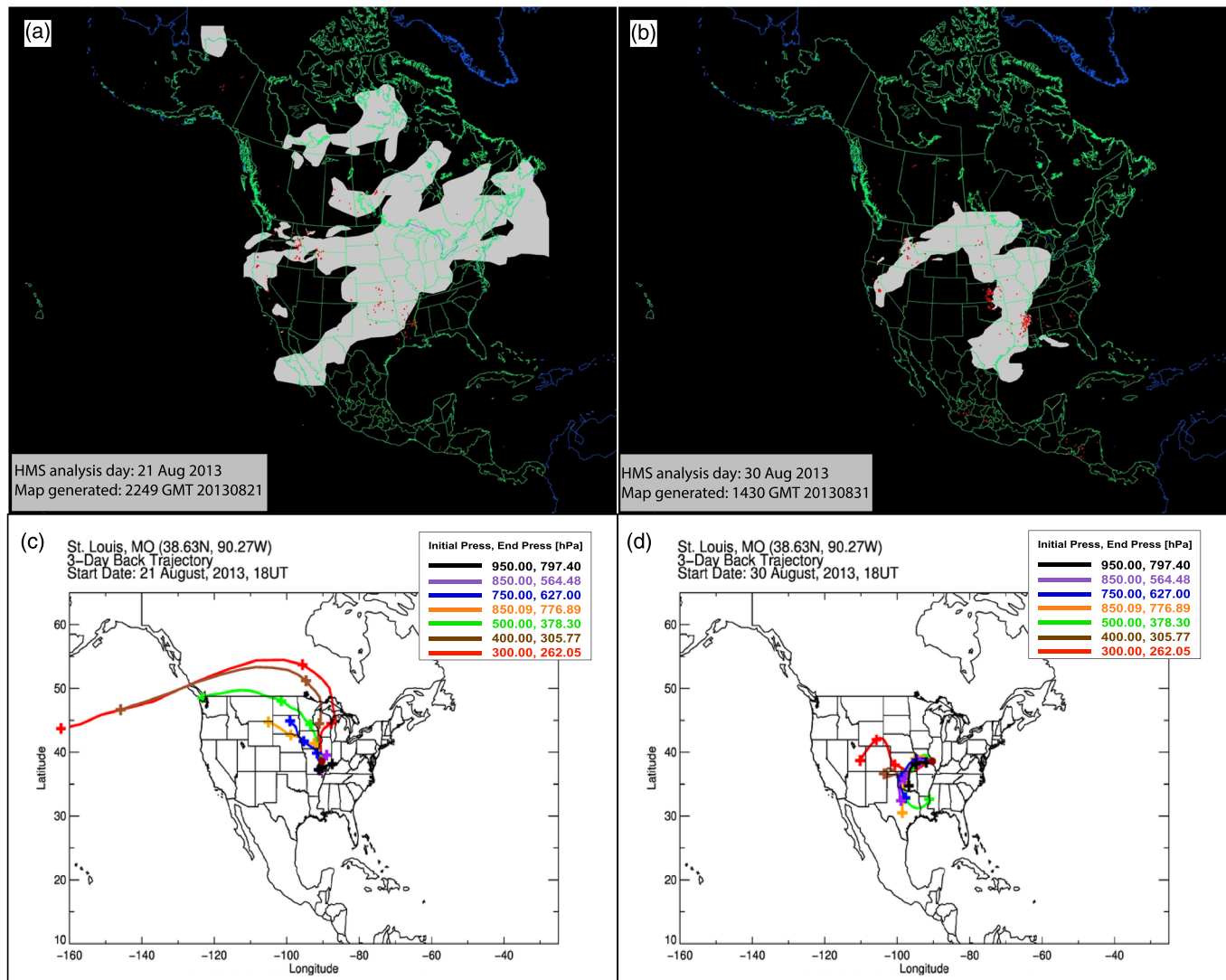
**Figure 6.** Series of daily synoptic atmospheric plane slices at 500 hPa at 7:00 UTC for preceding conditions to individual transport cases (a–c) 21 August 2013 and (d–f) 30 August 2013.



**Figure 7.** Atmospheric plane slices from NARR at 500 hPa at 18:00 UTC (a) 21 August 2013 and (b) 30 August 2013. Vertical cross sections at 90°W, 20–60°N 18:00 UTC (c) 21 August 2013 and (d) 30 August 2013. Blue contour lines show potential vorticity (PV)  $10^6$  PVU; relative humidity (RH) below 30% is shaded from light to dark, with darker shades representing the dryer air. The black lines on (a) and (b) are pressure height contours in m. While, the black lines on (c) and (d) represent potential temperature (isentropic surfaces) in K. Biomass burning sources tend to have low PV and can have moderate RH levels for pyroCbs, while stratospheric air masses have high PV and low RH. The blue dashed line PV contour (1.5 PV) shows the approximate boundary between stratospheric and tropospheric air masses. Arrows (green) show air mass transport patterns from stratospheric-to-tropospheric transport (STT) and from biomass burning (BB).

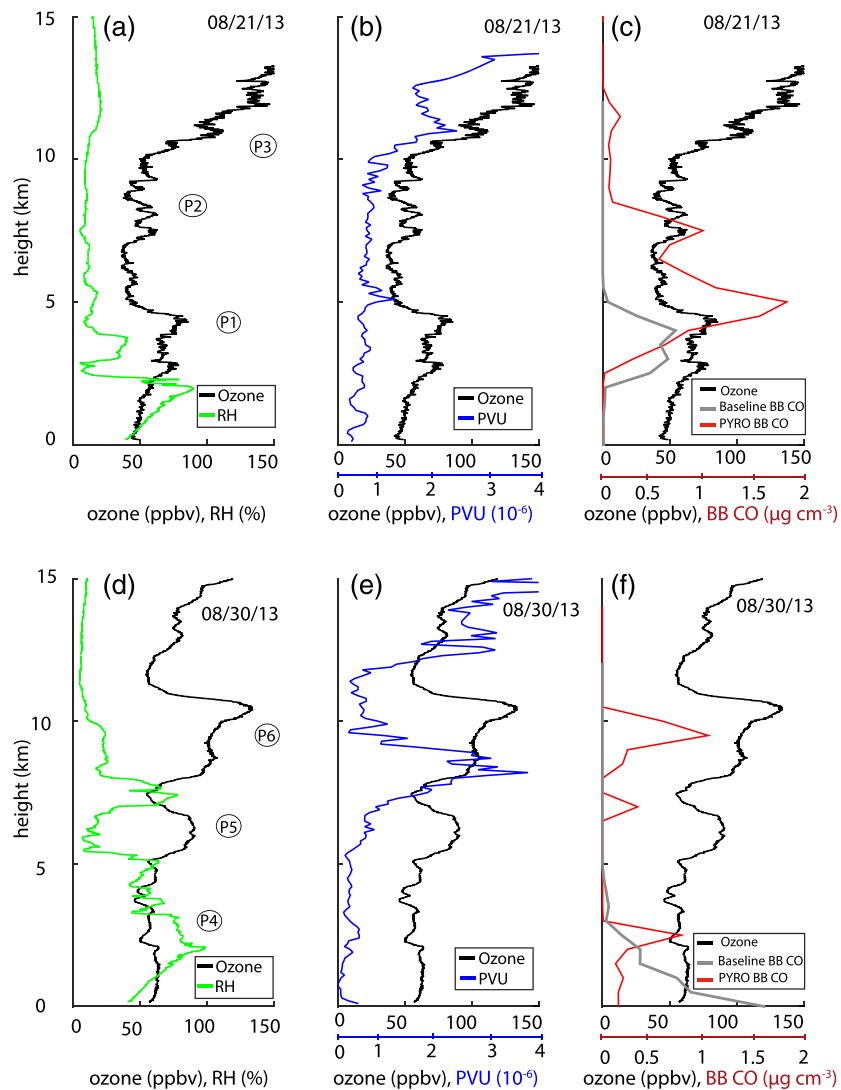
stratospheric air and ascending moist tropospheric air. The near surface plume (0 to 3.5 km) was traced back to southeastern agricultural fires ~5 days prior (see Figure 8), indicating a likely recirculation of air by the high pressure system. The middle-tropospheric plume P5 was determined to be from an unknown source, potentially a recirculation of summertime ozone precursors (Cooper et al., 2006, 2007). Evidence of this layer is present in previous soundings for the week 25–29 August where all sondes show a single well-defined





**Figure 8.** NOAA Hazard Management System (HMS) and Smoke Product analysis of smoke, (a) 21 August 2013 and (b) 30 August 2013. Below, 3-day backward FLEXPART-WRF trajectories corresponding to each episode is provided, (c) 21 August 2013 and (d) 30 August 2013. Additional trajectories are placed in the data archive online (at <https://tropo.gsfc.nasa.gov/seactions/>).

plume at ~5 to 9 km. The GEOS-5 and NARR potential vorticity also give no indication of what the cause for this layer is. Our hypothesis is supported by the vertical wind profiles for the week being relatively stagnant—the ozonesonde launched that day landed within 50 m of the launch site—signifying air circulation over St. Louis for the entire week increasing photochemical effects. An alternative theory is that wind shear caused the layer and it's a part of the larger plume above. The layer between the unknown plume and above (~6 to 8 km) is a layer of moist and clean air (see Figure 7d). The upper-tropospheric layer plume P6 is a combination of both a biomass burning plume and a stratospheric intrusion air mass. The lower half of the plume P6 (8 to 10 km) is dominated by stratospheric air, while the above portion of P6 (10 to 12 km) is primarily biomass burning. There is another layer of clean air above prior to reaching the tropopause. The upper-tropospheric plume P6 originated 5 days prior from high-altitude injection plumes from the California Rim Fire (25 August). The contribution of 30 August tropospheric ozone layers from biomass burning was 10 to 80 ppbv, and stratospheric air masses contributed 10 to 40 ppbv. The incorporation of high-altitude smoke injection allowed for the two middle- and upper-tropospheric polluted plumes to be characterized which contributed ~15% to 60% to layers in the total ozone column.



**Figure 9.** Vertical tropospheric profiles over St. Louis at ~18:30 to 19:30 UTC for selected test cases 21 August 2013 and 30 August 2013. Labels P1 to P6 correspond to plume information in Table 3. Ozone measured from the ozonesonde in ppb are shown as a black line. Panels (a) and (d), the green line RH %. Panels (b) and (e), the blue line represents the GEOS-5 modeled potential vorticity  $10^6$  PVU. Panels (c) and (f), the FLEXPART-WRF modeled CO biomass burning  $\text{g cm}^{-3}$ , for each simulation the PYRO simulation is red, and BASELINE simulation in gray. Refer to Figure 2 for corresponding ozonesonde curtain plots.

## 5. Summary and Conclusions

By incorporating balloon-borne ozonesonde observations with models, this study has quantitatively examined sources for tropospheric ozone enhancements due to NCOS. In particular, the pollution impacts from stratospheric intrusions and biomass burning contributions to background tropospheric and surface ozone levels in the Midwest United States were characterized. Additionally, emphasis is placed on partitioning the contribution from western U.S. fires, central U.S. fires, and other areas to St. Louis background ozone. A chemical transport model and trajectory model were run to quantify source contribution to ozone in St. Louis, Missouri. For the region and time period of this study, 10% to 15% of the ozone enhancements stems from a stratospheric air mass contribution and 15% to 30% from biomass burning. These NCOS contributions and ozonesonde profiles can be considered as baseline ranges for the Midwest U.S. area if direct ozone measurements (sondes, airplane, and ground based) are not available. Considering U.S. fires only, 70% of the biomass burning plumes originated from the western parts of the United States and only 3% came from the local

central U.S. emissions. Moreover, it was demonstrated that a redistribution of the biomass burning emissions injection height, with part of the emissions above the boundary layer, led to a reduction of model predicted surface ozone.

In agreement with earlier studies (e.g., Fishman et al., 2014), this study has identified a generally increasing relationship between background ozone and transported pollution. We followed the definition of background ozone described in Jaffe et al. (2018) as NCOS such as lingering biomass burning and long-range transported international sources. The major contributions below 3 km were from the central U.S. fires. While 80% to 90% of the high-altitude injection smoke (above 3.5 km) originated in the western United States. During this campaign period only 5% to 10% of the biomass burning emissions reaching St. Louis originated from the southeast and other regions.

This study identified that biomass burning plumes in the western United States can have impacts on the daily atmospheric ozone column in the Midwest (10 to 80 ppbv of ozone) at a greater frequency and intensity than stratospheric intrusion (10 to 25 ppbv of ozone). We show the background ozone to be 55 ppbv, which was near the 30- to 50-ppbv range mentioned in Jaffe et al. (2018) which is typical for the United States. We identified a relationship between smoke plume age and ozone enhancement where the high-altitude injection smoke plumes above 3.5 km generally were associated with higher amounts of CO concentrations but fresher smoke regarding ozone levels. In addition, we recognized that the high-altitude smoke had a higher tendency to mix with stratospheric intrusions, which together doubled the ozone enhancement in the tropospheric column.

An investigation of several individual smoke plumes has shown the importance of incorporating high-altitude smoke injection in model simulations in addition to ensuring that accurate biomass burning locations and temporal allocation of intensity are included in model emissions. Up to 60% of the smoke plume lies above 3.5 km, and this needs to be simulated as it can later be mixed down to the surface and lead to ozone exceedances days later. It was shown that model performance can be improved with the incorporation of satellite-based detections of high-altitude smoke injection (e.g., pyroCb activity, Peterson et al., 2014; Peterson, Fromm, et al., 2017; Peterson, Hyer, et al., 2017). In addition, modeling high-altitude smoke injection can help explain ozone enhancements aloft as well as at the surface.

The individual cases were selected because they are associated with common meteorological situations in the Midwest leading to ozone exceedances. Evidence from the first case where biomass burning is advected by a cutoff low is an example of a common flow pattern that transports air masses from the west. Summertime occurrences of this synoptic situation in conjunction with large wildfires in the western United States can lead to increases in ozone in the Midwest of 10 to 80 ppbv or greater. In the second case a stratospheric air mass was shown to mix with an aged wildfire plume during anticyclonic flow, a pattern that was previously found to occur 40% of the time for the southcentral United States (Texas and the Gulf of Mexico area, Brioude et al., 2007). Additionally, the test cases showed that the surface impacts were connected to mechanisms causing air parcels to move downward (e.g., shortwave on 30 August). Likewise, strong vertical motion was evident in bringing simulated air within the boundary to the middle and upper portions of the troposphere.

While the results in this study highlight that NCOS can contribute significantly to local tropospheric ozone in the Midwest, future studies must combine satellite data and model integration techniques with meteorological information for a longer time period, both within and outside the Midwest to better characterize NCOS contribution to U.S. ozone. Good examples of how an air quality model could be used to assess NCOS have been shown in Baker et al. (2016, 2018). More specifically, Baker et al. (2016, 2018) address the question of regional-scale pyrogenic ozone sources. In particular, Baker et al. (2018) investigated the Rim Fire, using non-sonde data, which occurred during our study period with a photochemical model, and their findings were similar to ours. Furthermore, as shown by individual plume cases and several earlier studies (e.g., Brioude et al., 2007; Hess & Zbinden, 2013; Jaffe, 2011; Lin et al., 2012; Morris et al., 2006), high-altitude smoke injection from a fire can lead to long-distance transport depending on weather conditions and hence impact surface ozone and NAAQS attainment. Plume rise and plume injection heights are a key source of uncertainty (Paugam et al., 2016), which can be reduced considerably using plume height information from remote sensing tools (e.g., Peterson, Fromm, et al., 2017; Peterson, Hyer, et al., 2017; Val

Martin et al., 2018). Improved modeling techniques will be required to better characterize biomass burning transport and hence better simulate long-range impacts and the possibility of unhealthy surface concentrations far from the biomass burning sources.

## Data Availability Statement

The data sets used in this work are publicly accessible and archived at <https://tropo.gsfc.nasa.gov/seacions/> (ozonesonde data and trajectories) and [https://www.nasa.gov/mission\\_pages/seac4rs/index.html](https://www.nasa.gov/mission_pages/seac4rs/index.html) (mission data, e.g., fire emissions and flight information).

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