

High Temporal Resolution Satellite Observations of Fire Radiative Power Reveal Link Between Fire Behavior and Aerosol and Gas Emissions

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November 16, 2022

Abstract

Wildfire smoke influences on air quality and atmospheric chemistry have been underscored by the increasing fire prevalence in recent years, and yet, the connection between fire, smoke emissions, and the subsequent transformation of this smoke in the atmosphere remains poorly constrained. Toward improving these linkages, we present a new method for coupling high-time-resolution satellite observations of fire radiative power (FRP) with in situ observations of smoke aerosols and trace gases. We apply this technique to thirteen fire plumes comprehensively characterized during the recent FIREX-AQ mission and show that changes in FRP directly translate into changes in conserved smoke tracers (CO₂, CO, and black carbon aerosol) observed in the downwind smoke plume. The correlation is particularly strong for CO₂ (mean $r > 0.9$). This method is important for untangling the competing effects of changing fire behavior versus the influence of dilution and atmospheric processing on the down-wind evolution of measured smoke properties.

	Grass / Shrubland	Grass / Shrubland	Grass / Shrubland	Grass / Shrubland	Forest / Woodland	Forest / Woodland	Forest / Woodland	Forest / Woodland	Mixed	Mixed	Mixed	M
Fire	Tucker	Castle	Castle	Sheridan	Shady	Left Hand	Lick Creek	Horsefly	North Hills	Ridgetop	Williams Flats	V

	Grass / Shrubland	Grass / Shrubland	Grass / Shrubland	Grass / Shrubland	Forest / Woodland	Forest / Woodland	Forest / Woodland	Forest / Woodland	Mixed	Mixed	Mixed	M
Fire Danger	Very High	Moderate	High	High	High	Moderate	High	High	High	High	High	V
Date Sampled	07/29	08/12	08/13	08/16	07/25	07/30	08/02	08/06	07/29	08/02	08/03	0
# Transects	10	15	23	18	43	12	6	10	13	9	32	7
# Detections	914	180	29	460	367	142	184	181	123	617	1162	1
Smoke Age	136 ± 76	176 ± 126	102 ± 54	165 ± 111	109 ± 56	334 ± 84	74 ± 44	116 ± 50	101 ± 74	150 ± 55	178 ± 95	8 4
r (CO ₂ :FRP)	0.98	0.95	0.95	0.99	0.99	0.99	0.99	0.99	0.80	0.99	0.99	0
r (CO:FRP)	0.98	0.78	0.62	0.87	0.66	0.99	0.99	0.99	0.80	0.96	0.51	0
r (BC:FRP)	0.97	0.37	0.36	0.60	0.81	0.99	0.99	0.79	0.79	0.88	0.67	0
r (MCE:FRP)	0.98	0.95	0.94	0.99	0.99	0.84	0.99	0.99	0.80	0.99	0.99	0

Table 1: Correlations between the relative rate of change in in situ measurements and GOES FRP. Fires are grouped into their dominant landcover type determined using final GeoMAC burned area perimeters and FCCS fuel maps. Observed fire danger, date sampled by the DC-8, the total number of transects per fire, the total number of daily GOES detections, and the average and standard deviation of sampled smoke age (min) is shown in the upper two panels. The third panel shows Pearson’s correlation coefficient (r) between the relative rate of change in transect integrated measurements $\left(\frac{\partial \ln(X)}{\partial t}\right)$ and the relative rate of change in FRP $\left(\frac{\partial \ln(FRP)}{\partial t}\right)$ integrated over the same time interval. Significant correlations (p-value < 0.005) are bold and insignificant correlations are not. Red grids show strong correlations ($r > 0.8$), orange grids show moderate correlations ($0.8 > r > 0.5$, and blue grids show weak correlations ($r < 0.5$).

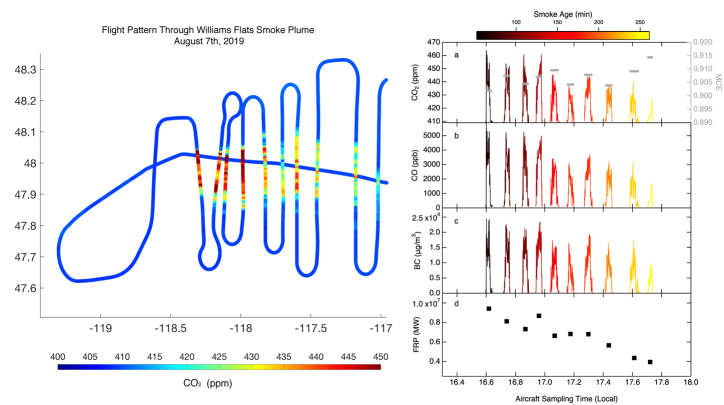


Figure 2: Left figure shows a map of the DC-8 flight track on August 7th, 2019 for the first set of orthogonal transects through the Williams Flats smoke plume. Colors correspond to the CO₂ mixing ratios from DC-8 measurements. Right figure shows a time series of CO₂, CO, and BC observations from the DC-8 (panels a-c) that correspond to the transects shown in the left panel and are highlighted by average smoke age. Panel d shows GOES FRP integrated over the same time interval represented by the smoke plume transects and aligned in time with the observations.

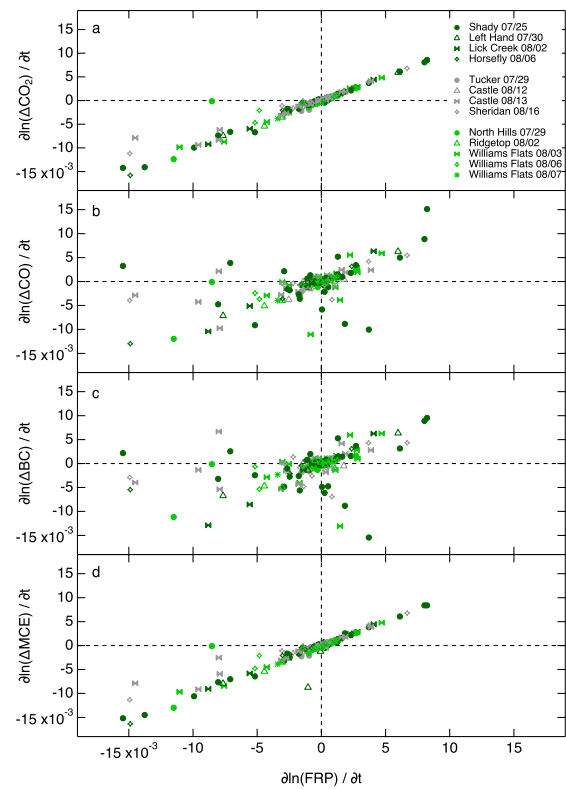


Figure 3: The relative rate of change in CO_2 (a), CO (b), BC (c), and MCE (d) versus the time aligned relative rate of change in GOES FRP. Colors are used to distinguish landcover types with light green representing a mixture of grass/shrubland and forest/woodland, dark green: forests / woodlands, and grey: grass/shrubland. Dotted black lines show zero change for the x and y-axis as a reference.

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Key Points:

- Geostationary satellite observations of fire radiative power are highly correlated with in-situ airborne measurements of primary-emission smoke tracers
- High resolution satellite observations are needed to disentangle how fire activity and plume dilution impact the downwind evolution of smoke
- Diurnal fire activity for wildfires observed during FIREX-AQ is best parameterized using a bimodal Gaussian distribution to inform models

Key Words: FIREX-AQ, fire, aerosol, smoke, black carbon, fire radiative power, emissions, transport, dilution, in-situ, trace gas

Abstract

Wildfire smoke influences on air quality and atmospheric chemistry have been underscored by the increasing fire prevalence in recent years, and yet, the connection between fire, smoke emissions, and the subsequent transformation of this smoke in the atmosphere remains poorly constrained. Toward improving these linkages, we present a new method for coupling high-time-resolution satellite observations of fire radiative power (FRP) with in situ observations of smoke aerosols and trace gases. We apply this technique to thirteen fire plumes comprehensively characterized during the recent FIREX-AQ mission and show that changes in FRP directly translate into changes in conserved smoke tracers (CO_2 , CO, and black carbon aerosol) observed in the downwind smoke plume. The correlation is particularly strong for CO_2 (mean $r > 0.9$). This method is important for untangling the competing effects of changing fire behavior versus the influence of dilution and atmospheric processing on the down-wind evolution of measured smoke properties.

1 Introduction

Wildfire activity in the western United States causes poor air quality, adverse human health impacts, and substantial economic costs (Jaffe et al., 2008; Kochi et al., 2010; Liu et al., 2015; Lu et al., 2016; Reid et al., 2016; Stavros et al., 2014). The frequency and intensity of these fires are expected to increase in the future due to a combination of growing human settlement at the wildland urban interface and climate change (Abatzoglou & Williams, 2016; Hammer et al., 2009; Mell et al., 2010; Theobald & Romme, 2007; Westerling et al., 2006). Consequently, it is essential to understand the composition and magnitude of aerosol and trace gas emissions from wildland fires and prescribed fires to quantify the effects of fire emissions on air quality and climate.

Fires emit a complex and highly variable mixture of gases and aerosols that can considerably alter atmospheric composition and tropospheric chemistry over a wide range of spatial and temporal scales (Bond et al., 2013; Goldammer et al., 2008; Langmann et al., 2009; Urbanski, 2014). Environmental conditions at the location of the fire such as local weather and fuel structure influence the composition and magnitude of these emissions (Loehman et al., 2014; Thonicke et al., 2010). Wildfires generally have a pronounced diurnal cycle directly related to weather conditions, with activity peaking early in the afternoon and diminishing after

sunset (Ichoku et al., 2008; Saide et al., 2015; Zhang & Kondragunta, 2008).

Fire emissions inventories are an essential tool for understanding the spatio-temporal distribution of fire emissions on a regional to global scale. The resolution of commonly used fire emissions inventories diverges considerably depending on their intended use and the methodology used in their development. As a result, considerable irregularities exist among fire emissions inventories in the estimated magnitude, composition, and distribution of emissions in space and time (Larkin et al., 2014; Li et al., 2019a; Liu et al., 2020; Shi et al., 2015). In general, these differences can be attributed to variations in the approach used to quantify burned area, fuel loads, combustion completeness, and emission factors (Kasischke & Penner, 2004). Quantitative comparisons between different fire emissions inventories remain difficult due to the variable transport models used in each study and the spatial/temporal averaging used for comparison to observations (Liu et al., 2020).

Most fire emissions inventories employ remote sensing observations of fire parameters such as burned area, active fire counts, and fire radiative power (FRP) from instruments onboard polar orbiting satellites, including the Moderate Resolution Imaging Spectroradiometer (MODIS) (Ichoku & Ellison, 2014; Kaiser et al., 2012; van der Werf et al., 2017; Wiedinmyer et al., 2011, Pierce et al., 2007) and the Visible Infrared Imaging Radiometer Suite (VIIRS) (Ahmadov et al., 2017). Typical overpass times for the satellites hosting these instruments occur only twice daily over North America at ~1am/pm or at ~10am/pm local time (Li et al., 2018). Due to this limited temporal coverage of fire observations in a given location, some fire emissions inventories or models supplement the diurnal cycle of emissions using FRP observations from geostationary satellites (Andela et al., 2015; Li et al., 2019b; Mota & Wooster, 2018; Mu et al., 2011; Zhang et al., 2012), or assume a Gaussian distribution of daily FRP (Kaiser et al., 2009).

Geostationary satellite instruments such as the Geostationary Operational Environmental Satellite (GOES) Advanced Baseline Imager (ABI) observe fire radiative power at a higher temporal resolution than their polar-orbiting counterparts and over the complete diurnal cycle, but this comes at the expense of hemispheric, not global coverage and detection biases due to lower spatial resolution since errors generally increase with decreasing fire size and radiative power (Li et al., 2020; Schmidt, 2019). GOES ABI imagery provides a snapshot of FRP across the continental United States every 5 minutes and full disk FRP every 10 minutes (<https://www.goes-r.gov/spacesegment/abi.html>) at a relatively coarse spatial resolution of 2 km

(Schmidt, 2019), and offers the opportunity to investigate both the diurnal cycle of fire activity and short-term changes in fire behavior that could have important implications for fire emission estimates (Li et al., 2019b; Schmidt, 2019; Zhang & Kondragunta, 2008).

There is a need to connect spatially-coarse, remotely-sensed fire observations that have more widespread coverage and lower time resolution with in-situ point source observations that have much higher spatial and temporal resolution but much lower overall coverage to achieve a more comprehensive understanding of fire behavior and emissions. Rather than using GOES FRP observations to directly estimate emissions using a top-down approach (Freeborn et al., 2008; Ichoku et al., 2008; Wooster et al., 2003), we use 5-minute resolution, near real time GOES FRP observations to quantify the relationship between FRP and in situ measurements of fire emissions. We validate this technique using airborne observations of individual fires sampled during the recent Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ) field campaign. FIREX-AQ measured the concentrations, composition, and properties of smoke from wildfires and prescribed fires in the Western United States during the summer of 2019 using the NASA DC-8 aircraft. The GOES FRP observations are compared to the airborne in situ measurements of relatively long-lived species emitted by fires, including CO₂, CO, and black carbon (BC) mass. This work is important and relevant for new and existing daily fire emissions inventories or smoke forecasting model frameworks and promises significant improvements for resolving the temporal resolution of emissions.

2 Methods

2.1 GOES FRP

2.1.1 GOES Diurnal Cycle of FRP

FRP is an important quantitative indicator of fire activity and how it changes over the study period. We generate an average diurnal cycle of FRP for all the Western US wildland fires in FIREX-AQ by examining each fire sampled individually using FRP observations from the GOES-16 and 17 Advanced Baseline Imager (ABI) L2+ Fire/hot spot Detection and Characterization (FDC) product from the Wildfire Automated Biomass Burning Algorithm (WFABBA) processing system (Schmidt, 2019). The FRP data products for the wildland fires specific to FIREX-AQ are archived in the NASA data archive. This product provides FRP for

fires in the continental US with a spatial resolution of 2 km and a temporal resolution of 5 min. Previous studies have found the diurnal cycle of fire activity can be modeled as a Gaussian distribution or a Fourier series (Andela et al., 2015; Giglio, 2007; Roberts et al., 2009; Zhang & Kondragunta, 2008). In this study, we choose to use a bimodal Gaussian distribution as opposed to one with a single mode, because the bimodal fit had a slightly higher correlation with the observation-driven diurnal cycle of FRP for the FIREX-AQ fires (Supplementary Table 1). For each individual fire, we derive the average diurnal cycle of FRP, defined as the fit to the total FRP (in 5-min intervals over the course of a 24-hour day in local time) averaged over every day the fire was actively burning (Figure 1). A mean diurnal cycle for multi-day fires is used to fill in observational data gaps when cloud cover prohibits detection. The functional form of the bimodal Gaussian distribution is

$$\text{FRP}(t) = \sum_{i=1}^2 \frac{a_i}{\sigma_i \sqrt{2\pi}} e^{-\left(\frac{t-t_i}{\sigma_i \sqrt{2}}\right)^2} \quad (1)$$

where a_i is the FRP of mode i in units of MW, t_i is the median time of mode i in hours from midnight (local time) and σ_i is the standard deviation of mode i in hours. GOES FRP observations are included if their centroid is within 4km of the final fire perimeter defined by the Geospatial Multi-Agency Coordination (GeoMAC) Wildland Fire Perimeters database (Walters et al., 2011). The size of the buffer is double the diameter of the fixed grid resolution from GOES in order to capture all detections of an individual fire.

To quantify the diurnal cycle of FRP for a single fire on an individual day, any time-gaps in GOES FRP observations were filled using the average Gaussian model scaled to the mean of the available FRP observations for each fire on that day. Observational gaps often occur because of cloud cover, and on average this occurred 34% of the time for the fires included in this analysis. The model fit is only used to interpolate missing FRP observations for days with observations of FRP > 0 spanning at least 6 hours out of the entire 24-hour (local time) period in order to avoid overestimating FRP. In practice, this data reconstruction step has negligible impact on the FRP timeseries for fires where data coverage is good (e.g., the Williams Flats fire highlighted in Figures 2 and S1); however, for the few fires with substantial missing observations due to cloud cover (e.g., Castle fire), this step is more impactful.

2.1.2 Fuels and Fire Weather

We investigate the influence of fuel type on the mean diurnal cycle of FRP by grouping fires that burned in ecosystems with similar fuel loading and structure, using the Fuel Characteristics and Classification System (FCCS) (Ottmar et al., 2007) to determine the dominant ecosystem type that was consumed. The dominant ecosystem type is defined as the FCCS fuel class that encompasses at least 75% of the burned area defined by the final GeoMAC perimeter at the end of the fire's lifecycle. It should be noted that only the dominant fuel type per fire was used for classification in this study, although each fire burned through a wide variety of fuel types, and different ratios of fuel types on different burn-days are neglected. The fires included in this analysis are grouped into one of the 3 following fuel categories: grass/shrublands, forest/woodlands, and mixed. The mixed category is defined as a combination of grass/shrublands and forest/woodlands for fires with less than 75% of the burned area encompassed by either grass/shrubland or forest/woodlands. Daily fire weather danger levels are obtained from the National Fire Danger Rating System provided by the US Forest Service (Bradshaw et al., 1983).

Fires in grass / shrublands are often considered to be fuel limited, while forest fires can be considered ignition limited because of generally higher fuel moisture. The diurnal cycle of fire activity begins around the same time for all ecosystems considered (12:00 hours local time), but extended much later into the night (24:00 hours) in mixed ecosystems compared to grass / shrublands or forest / woodlands (20:00 hours), with the exception of the Horsefly fire (Figure 1). Incident reports of Horsefly indicate the fuels include a significant amount of dead and down trees caused by bark beetle damage (<https://inciweb.nwcg.gov/incident/6502/>). The longest diurnal combustion period occurs in mixed fuels and forests with high proportions of beetle killed dead and down trees. One explanation for this behavior is the finer fuels allowed for more rapid fire spread and helped to dry out the larger dead fuels. Simultaneously, smoldering combustion in coarse woody debris, including beetle killed trees, is known to continue well into the night (Albini & Reinhardt, 1995; Hyde et al., 2011). In all of the fires categorized as having mixed fuels, the invasive species cheatgrass was a component of the fuels in the burned area perimeter (24% Williams Flats, 12% North Hills, and 4% Ridgetop). Cheatgrass enhances fire size and frequency in the Western US and can outcompete native vegetation after fire (Balch et al., 2013; Kerns et al., 2020; Menakis et al., 2003).

2.2 Comparison with In situ Observations

2.2.1 Aircraft Aerosol and Trace Gas Observations

We compare the high-temporal resolution GOES FRP emission product to the airborne measurements of CO₂, CO, and refractory BC aerosol concentrations in the FIREX-AQ smoke plumes. The DC-8 aircraft flew through each wildfire smoke plume in a series of orthogonal plume transects starting near the fire and proceeding downwind as far as practical given mission objectives and flight limitations (Figure 2). An example set of plume transects for the Williams Flats fire in Washington State is shown in Figure 2. CO₂ mixing ratio measurements are obtained using a non-dispersive IR spectrometer (LICOR, Inc. Model 7000) adapted for aircraft measurements in a method similar to *Vay et al.* (2003), while CO mixing ratios are obtained from mid-IR laser absorption spectrometry (Sachse et al., 1991). Both species were calibrated in-flight with standards from NOAA ESRL traceable to WMO scales (CO₂:X2007; CO:X2014A). Refractory BC mass concentrations appropriate for most of the accumulation-mode were provided by a Single Particle Soot Photometer (SP2, Droplet Measurement Technologies). CO₂ is chosen because it is the most dominant trace gas species emitted from fires (Andreae & Merlet, 2001). CO and BC are chosen for comparison because they are conserved tracers of primarily smoldering and flaming fire processes (Sommers et al., 2014; Urbanski, 2014), respectively, over the relatively short (hours-long) timescales of the DC-8 sampling. The modified combustion efficiency (MCE), is a metric commonly used to determine relative contributions from smoldering and flaming fire processes to emissions (Ward & Radke, 1993). MCE is calculated using the following equation:

$$MCE = \frac{1}{m_{CO/CO_2} + 1} \quad (2)$$

where m_{CO/CO_2} is the slope of the York regression between excess mixing ratios (background subtracted) of CO and CO₂.

2.2.2 Relationships between In-Situ Measurements and GOES FRP

To compare in-situ trace gas and aerosol observations with GOES FRP, we calculate the smoke age as the difference between when the smoke was emitted and when it was sampled by the DC-8, using the aircraft-measured wind speeds and assuming straight line horizontal advection between the fire and aircraft positions with uniform winds for all transects of a single plume. The average wind speed for all fires and all transects is $8 \pm 3 \text{ m s}^{-1}$ and the typical wind

direction is westerly. The vertical transport time of the plume is neglected. We calculate the time of emission as the average time of sampling by the DC-8 aircraft across a single transect minus this smoke age.

We quantify the temporal variability of CO₂ and CO mixing ratios as well as the BC mass concentration by integrating the DC-8 measurements across each smoke plume transect following the methodology of *Yokelson et al. (2007)*. For each orthogonal transect through the FIREX-AQ smoke plume, we integrate excess mixing ratios of CO₂, CO, and BC across the entire length of the plume cross-section. Baseline concentration values used for the background subtraction are calculated as the 5-second-averaged mixing ratios of each species starting one second before and after each transect. We then calculate the relative rate of change of the aerosol and trace gas species and MCE for each transect, which we expect should scale proportionately with the rate of change of the FRP, after accounting for the smoke plume age, Δt , as follows:

$$\left. \frac{\partial \ln(\Delta X)}{\partial t} \right|_{t+\Delta t} \propto \left. \frac{\partial \ln(\text{FRP})}{\partial t} \right|_t \quad (3)$$

where ΔX is the integrated excess mixing ratio of species X , and t is the time corresponding to the measured FRP and estimated time of smoke emission. In this study, we compute the approximate derivative by differencing the DC-8 measurements across two adjacent aircraft transects. For each DC-8 plume transect, k , (and corresponding time interval at the fire, j), the scaling relationship is calculated as

$$\frac{1}{\Delta X_k} \cdot \frac{\Delta X_k - \Delta X_{k-1}}{t_k - t_{k-1}} \propto \frac{1}{\Delta \text{FRP}_j} \cdot \frac{\Delta \text{FRP}_j - \Delta \text{FRP}_{j-1}}{t_j - t_{j-1}} \quad (4)$$

where ΔFRP_i is the integrated FRP over the relatively short time interval represented by the aircraft sampling time minus the smoke age ($t_j = t_k - \Delta t_k$). Implicit in Equation 4 is that the smoke age can be used to extrapolate from the aircraft sampling time back to the time of emission at the fire. Furthermore, we assume that the conserved species in the plume do not continue to evolve between adjacent transects (i.e., $\Delta t_k - \Delta t_{k-1} = 0$). While this assumption may be reasonably valid for the conserved tracers examined here whose plume evolution is mainly impacted by dilution, it is likely to break down for other extensive aerosol and trace gas variables that are strongly influenced by photochemical processing, coagulation, and gas-particle partitioning of semi-volatile compounds. Applying Equation 3 for those variables for FIREX-AQ will be more

complicated, as the spacing of the DC-8 sampling transects along the plume length do not reflect a 1:1 increase in both smoke age and sampling time interval. Figure S3 demonstrates that this ratio varies from 0.8-6.4 across the FIREX-AQ wildfires. In most cases, the time it takes the aircraft to sample successive downwind portions of the plume is considerably shorter than the time it takes for the plume to be advected over the intervening distance, assuming straight-line, horizontal advection.

We compute Pearson's correlation coefficients to quantify the linear proportionality between transect-integrated values of CO₂, CO, and BC versus FRP as represented by Equation 4. While strong correlation coefficients would be hypothesized to indicate the governing influence of fire activity on emissions, weaker correlation coefficients may indicate the presence of important, confounding processes such as smoke plume dilution. The importance of dilution for driving smoke variability may also vary at specific locations within the plume (e.g., near the edges versus the center) in ways that are not captured by this integrated plume analysis. Similarly, the nature of the aircraft horizontal sampling transects prevent us from examining changes in the vertical structure of these conserved tracer species that may be impacted by boundary layer convective mixing, dilution, or size-dependent particle gravitational settling.

3 Results and Discussion

3.1 GOES FRP Diurnal Cycles

We investigate the average diurnal cycle of FRP on the scale of individual fires in the western US grouped according to the dominant ecosystem represented by the burned area (Figure 1; Supplementary Table 1). The diurnal cycle of FRP for all fires in this analysis is optimally fit using a bimodal Gaussian distribution. Coefficients of the bimodal Gaussian distribution for all fires are tabulated in Supplementary Table 1. Pearson's correlation coefficients (r) between single-mode Gaussian distributions and bimodal Gaussian distributions (Supplementary Table 1) demonstrate the ability to model diurnal fire activity and highlight the slight differences between single and bimodal distributions.

Our results suggest a bimodal Gaussian distribution could improve the accuracy of the timing of fire emissions in fire emissions inventories and smoke forecasting models. The fires tend to peak later in the day than might be expected based on past literature (Giglio, 2007; Pack et al., 2000; Roberts et al., 2009; Zhang & Kondragunta, 2008), but the model standard

deviations of 1-2 hours are consistent with common model assumptions. We find the timing and magnitude of peaks in the bimodal Gaussian fit of the diurnal distribution of FRP vary between all fires, but there are discernible differences between the ecosystems represented by the fires analyzed. Overall, the 5-minute data reveal significant time-varying structure in fire activity, which needs to be accounted for when examining the fire plume characteristics across different aging timescales.

3.2 Relationships between Integrated GOES FRP and In Situ Measurements

Our approach allows for direct comparison of satellite FRP observations with 1 Hz aircraft trace gas and aerosol observations at an extremely high time resolution of 5-minute intervals. The strong linear relationship between fire-integrated FRP and the combustion rate of biomass is well established in the literature and is the basis for top-down fire emissions inventories (Freeborn et al., 2008; Ichoku et al., 2008; Wooster et al., 2003). Figure 3 shows the relationships between the relative rate of change in transect-integrated CO₂, CO, BC, and MCE and the corresponding relative rate of change in integrated FRP observations from GOES.

It is clear that the DC-8 was able to sample fire emissions from periods when the fire activity was both waxing ($\partial \ln(\text{FRP})/\partial t > 0$) and waning ($\partial \ln(\text{FRP})/\partial t < 0$), respectively. While many fires were sampled during periods of increasing fire activity where both increasing FRP and plume dilution serve to reduce concentrations of conserved tracers during downwind flight legs (relative to the earlier legs), some fires (e.g., Shady on July 25th and Sheridan on August 16th) exhibited a decrease in FRP over time. An example of a fire sampled during periods of increasing FRP is given in Figure 2 for Williams Flats fire, and a counterexample of a fire sampled during decreasing FRP is shown in Supplementary Figure 2 for the Sheridan fire. The plume peak timeseries shown in Figures 2 and S2 highlight the importance of the FRP trend as a governing influence on plume evolution. In Figure 2, the decrease in peak areas with increasing downwind distance is characteristic of plume dilution, while the lack of a decrease shown in Figure S2 implies that the plume is not diluting. It is only with the important context provided by the FRP timeseries that changing fire activity, rather than dilution, be considered as the primary driver of these starkly contrasting plume trends.

We uncover strong linear correlations ($r > 0.8$) between the relative rate of change in $\ln(\text{FRP})$ and the relative rate of change in both $\ln(\text{CO}_2)$ and $\ln(\text{MCE})$ (Table 1). Figure 3

highlights the exceptionally strong correlation between the rate at which FRP changes with time and the resulting relative temporal change in CO₂ mixing ratio observed by the DC-8 downwind (panel A). The strong correlations between FRP and CO₂ and MCE are likely because fire carbon emissions are composed of 80-90% CO₂ (Andreae & Merlet, 2001). The correlations remain strong ($r > 0.8$) on days with plentiful GOES detections to inform the diurnal cycle of FRP and also on days with scarce GOES detections when the Gaussian model fit to the average diurnal FRP cycle was relied on heavily. Smoke age did not have a discernible influence on the correlations over the range of FIREX-AQ variability (<6 hours old).

The correlation with the relative rate of change in FRP weakens slightly for CO and BC compared to CO₂. This may reflect the confounding influences of the fire properties on the emission of these incomplete combustion products (although not so much explained by the relative rate of change in MCE). While BC aerosols are also subject to plume processes such as coagulation that reduce their number concentration beyond what would be attributable to dilution with background air alone, the mass concentrations reported here should be largely conserved over the early hours of the plume. Limitations of this analysis include the lack of in-situ measurements that span the vertical length of the plume and the potential of horizontal heterogeneity in the distribution of emissions in the plume. LIDAR-derived measurements of vertical bulk aerosol extinction could offer an opportunity to explore the vertical distribution of emissions and the role of boundary layer dynamics on plume extent.

4 Summary and Conclusions

We present a new method for evaluating emissions inventories and the observed rates of change of conserved emissions tracers using high time-resolution satellite observations of FRP. The technique is used to interpret the comprehensive airborne dataset from the NASA FIREX-AQ mission in summer, 2019. These unique data demonstrate the need for and the power of satellite observations for disentangling the impacts of dilution, atmospheric processing and changing fire activity on fire emissions observed in smoke plumes. Our results suggest smoke forecast models could leverage assimilation of high-time resolution GOES FRP observations to significantly improve their ability to temporally distribute emissions. The strong relationships between the rate of change in FRP and CO₂ can also be exploited in smoke forecasting and emissions models, because it provides a connection to other trace gas and aerosol emissions. While fire emissions

are commonly modeled as a single, Gaussian mode, we show that this representation is oversimplified and would fail to capture the multi-peak structure of the diverse FIREX-AQ fires. The results from this study also imply that high-time resolution GOES FRP observations can be used as a tool to tease apart the influence of changing fire behavior from downwind plume processing when interpreting airborne campaign measurements. The variation in FRP over the time period represented by smoke plumes is an important factor in understanding smoke evolution along the length of a plume, and should be considered along with dilution and atmospheric processing. We demonstrate the strong connection between FRP and CO₂, which suggests that changes in fire activity govern the near-field plume concentrations more so than dilution. Combining airborne measurements with satellite FRP is a powerful analysis tool for accounting for the influence of changing fire activity on plume observations and their downwind evolution.

Acknowledgements and Data:

This work was supported by the NASA Tropospheric Chemistry Program managed by Dr. Barry Lefer. EBW is supported by a NASA Postdoctoral Program Fellowship. JMK and JPS are supported by NOAA. We thank the FIREX-AQ project scientists Jim Crawford, Carsten Warneke, and Jack Dibb, as well as the pilots and crew of the NASA DC-8. All data are publicly available from the NASA FIREX-AQ data archive at <https://doi.org/10.5067/suborbital/firexaq2019/data001>.

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High Temporal Resolution Satellite Observations of Fire Radiative Power Reveal Link Between Fire Behavior and Aerosol and Gas Emissions

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Contents of this file

Figures S1 to S3
Tables S

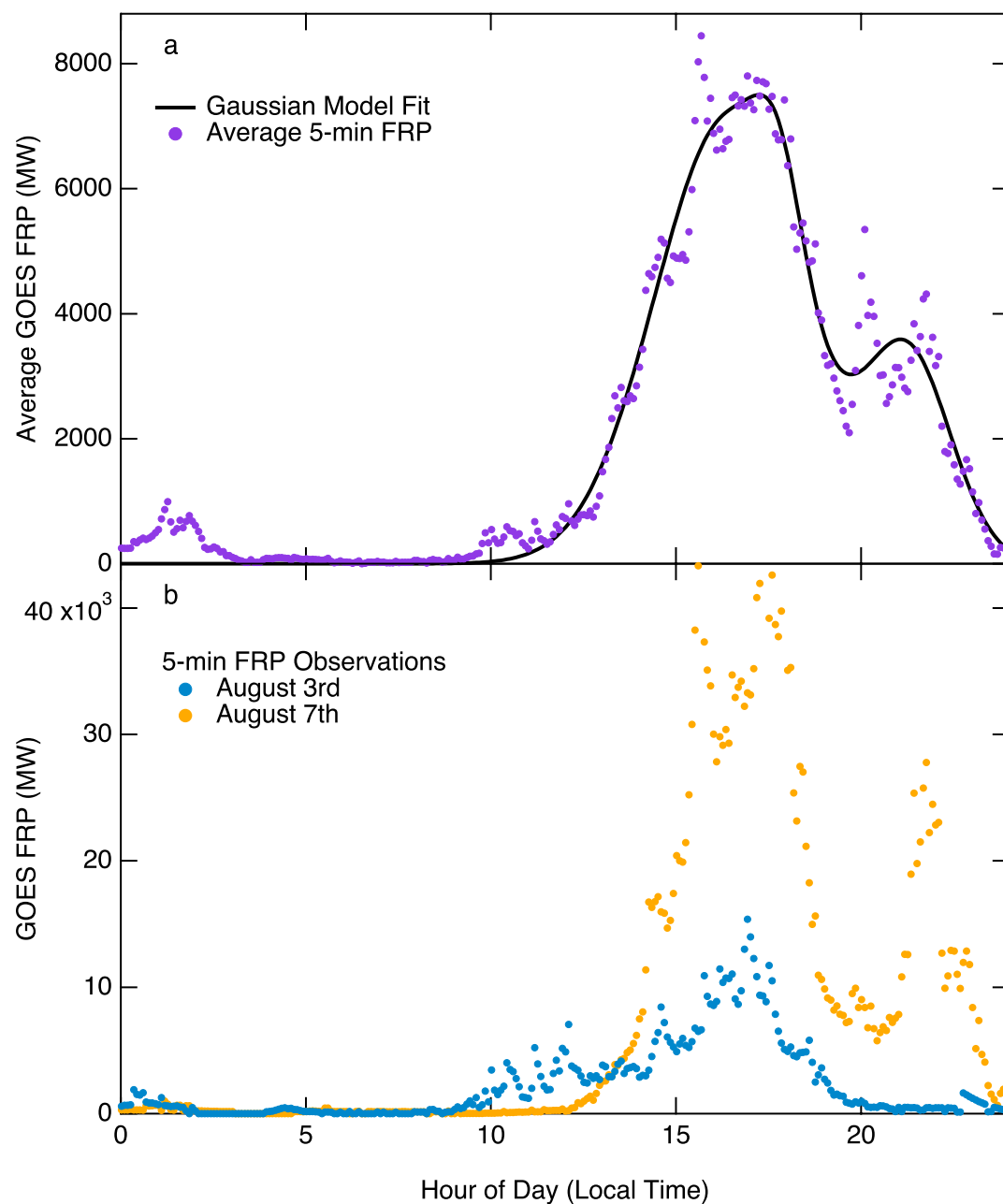


Figure S1. Panel a shows a time series in local time of the average GOES FRP diurnal cycle for Williams Flats fire over the entire lifetime of the fire. Average 5-min FRP is shown as purple circles. The FRP reflects the sum of all detections within a 5-min interval. The bimodal Gaussian model fit to the lifetime average 5-min FRP is shown as a black line. Panel b shows 5-min FRP for all observations on August 3rd (blue) and August 7th (gold). The Gaussian model fit shown in panel a is used to inform the interpolation of the daily 5-min FRP shown in panel b.

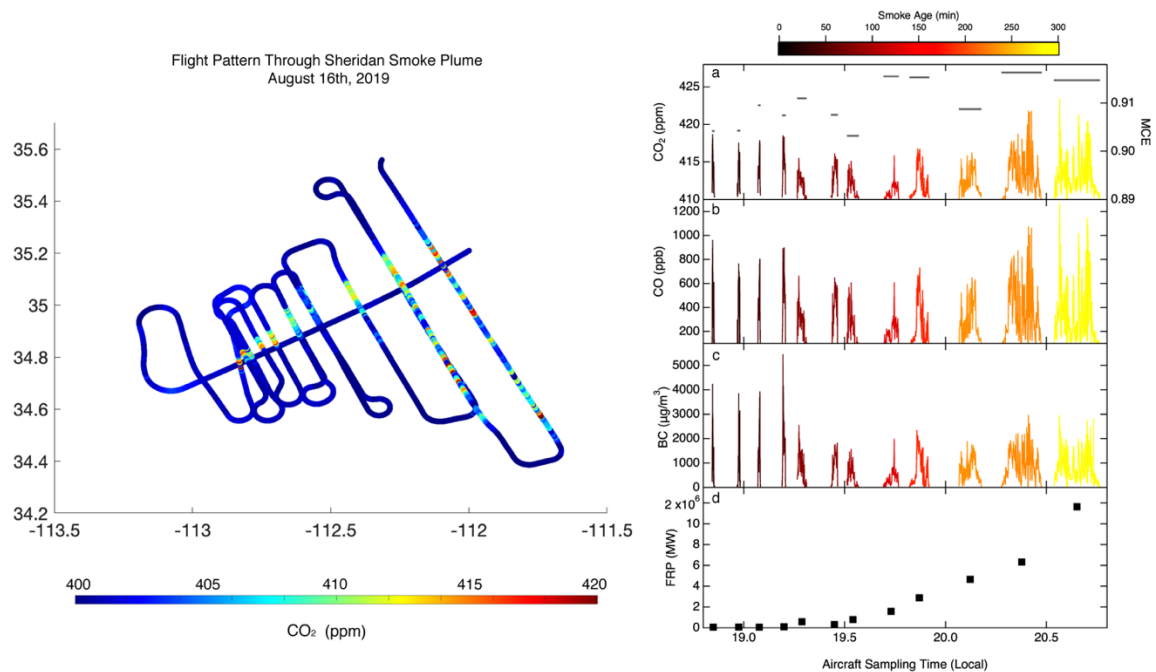


Figure S2. Left panel shows a map of the DC-8 flight track on August 16th, 2019 for the first set of orthogonal transects through the Sheridan smoke plume. Colors correspond to the CO₂ mixing ratios from DC-8 measurements. Example of positive rate of change in observations and corresponding FRP for Sheridan fire. Time series of CO₂, CO, and BC observations from the DC-8 (panels a-c) highlighted by average smoke age. Panel d shows the sum of GOES FRP integrated over the same time interval represented by the smoke plume transects and aligned in time with the observations.

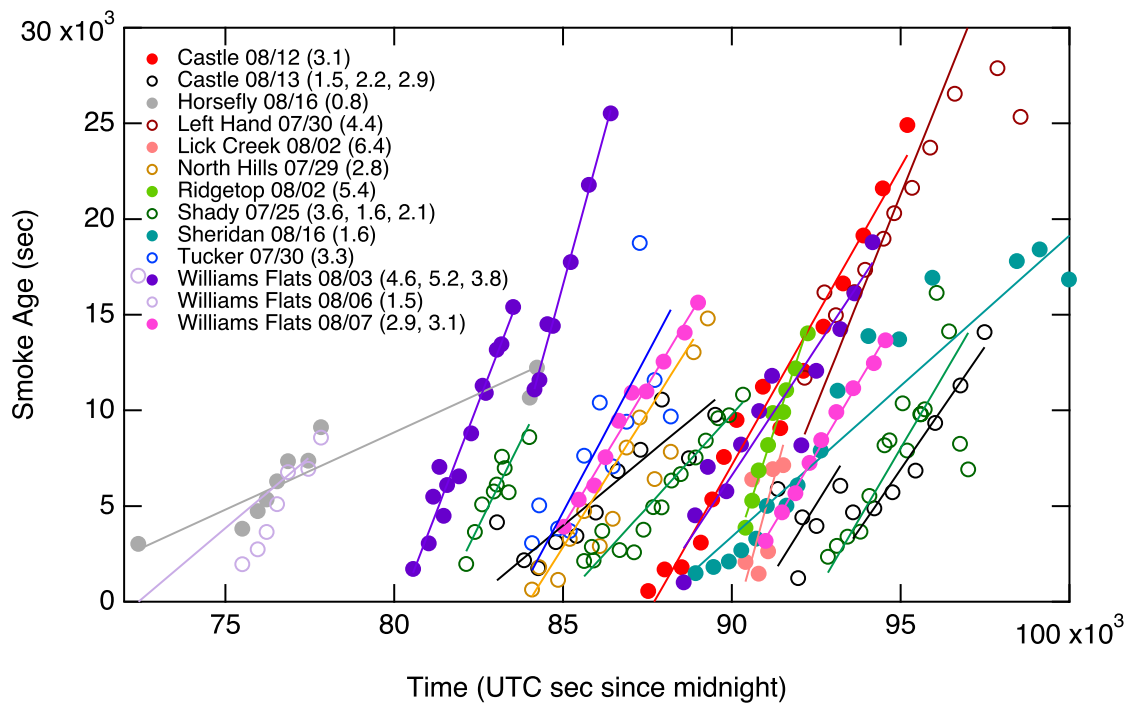


Figure S3. Relationship between time of sampling and smoke age. The linear fit is shown as solid lines and individual transects are shown as dots. Colors represent fires sampled on a given day. The slope of the linear fit for each fire is given in the legend following the fire name and sampling data in parenthesis. For days when the fire was sampled with more than one set of orthogonal transects going down the plume, the slope is given for all sets of transects sequentially.

	Grass / Shrublands			Forests / Woodlands				Mixed		
	Tucker	Castle	Sheridan	Shady	Left Hand	Lick Creek	Horsefly	North Hills	Ridgetop	Williams Flats
r (1)	0.91	0.97	0.95	0.89	0.91	0.86	0.76	0.76	0.84	0.95
r (2)	0.95	0.99	0.96	0.93	0.94	0.89	0.88	0.88	0.93	0.98
a ₁ (MW)	1623	117	917	372	158.5	242	256	617	947	3560
t ₁ (hr)	15.41	14.85	16.27	16.71	15.53	14.30	14.29	14.97	17.59	16.87
σ_1 (hr)	0.60	0.66	0.51	1.83	2.35	0.62	1.08	0.93	4.39	2.85
a ₂ (MW)	1074	60	1320	242	108	114	300	637	391	1611
t ₂ (hr)	17.98	16.54	17.07	19.87	18.75	15.66	17.04	20.05	18.45	21.896
σ_2 (hr)	1.68	2.041	2.34	1.01	5.39	0.54	0.94	2.57	0.60	1.692

Table S1: Fires classified into categories based on dominant landcover type of burned area. Pearson's correlation coefficient between the model fit and observation driven diurnal FRP cycle for single mode Gaussian distribution (r(1)) and bimodal Gaussian distribution (r(2)). Gaussian model fit parameters for the first mode (a₁), time of the first mode (t₁), and standard deviation of first mode (σ_1) are shown in the upper portion of the table. The bottom portion shows the Gaussian model fit parameters for the second mode, time, and standard deviation (a₂, t₂, σ_2).