

1 **Reconciling Assumptions in Bottom-up and Top-down Approaches for Estimating**
2 **Aerosol Emission Rates from Wildland Fires using Observations from FIREX-AQ**

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22 **Key Points:**

- 23 • In situ measurements of wildland fire smoke plumes provide emission rates for
24 evaluating emissions inventories at unprecedented resolution
- 25 • Fire emissions inventories struggle to capture the emissions rate characteristics of
26 individual fires but may perform well in the aggregate
- 27 • Bottom-up inventories suffer from major uncertainty in key variables, while top-down
28 inventories may have bias from imperfect assumptions
- 29

30 **Abstract**

31 Accurate fire emissions inventories are crucial to predict the impacts of wildland fires on air
32 quality and atmospheric composition. Two traditional approaches are widely used to calculate
33 fire emissions: a satellite-based top-down approach and a fuels-based bottom-up approach.
34 However, these methods often considerably disagree on the amount of particulate mass emitted
35 from fires. Previously available observational datasets tended to be sparse, and lacked the
36 statistics needed to resolve these methodological discrepancies. Here, we leverage the extensive
37 and comprehensive airborne in situ and remote sensing measurements of smoke plumes from the
38 recent Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ)
39 campaign to statistically assess the skill of the two traditional approaches. We use detailed
40 campaign observations to calculate and compare emission rates at an exceptionally high
41 resolution using three separate approaches: top-down, bottom-up, and a novel approach based
42 entirely on integrated airborne in situ measurements. We then compute the daily average of these
43 high-resolution estimates and compare with estimates from lower resolution, global top-down
44 and bottom-up inventories. We uncover strong, linear relationships between all of the high-
45 resolution emission rate estimates in aggregate, however no single approach is capable of
46 capturing the emission characteristics of every fire. Global inventory emission rate estimates
47 exhibited weaker correlations with the high-resolution approaches and displayed evidence of
48 systematic bias. The disparity between the low resolution global inventories and the high
49 resolution approaches is likely caused by high levels of uncertainty in essential variables used in
50 bottom-up inventories and imperfect assumptions in top-down inventories.

51 **Plain Language Summary**

52 Smoke emitted by wildland fires is dangerous to human health and contributes to climate change.
53 To predict and evaluate the impacts of fires, we need to know how much smoke is emitted into
54 the atmosphere. There are two state-of-the-art methods used to estimate the mass of smoke
55 emitted by fires, but they often disagree. In this study, we use unusually detailed measurements
56 collected using an aircraft that flew within wildland fire smoke plumes to calculate the amount of
57 smoke emitted from fires in the Western United States. We compare emission rates derived from
58 the exceptionally high spatial and temporal resolution approach to the two traditional, lower
59 resolution approaches to understand why they sometimes diverge.

60 **1 Introduction**

61 Wildland fires can be dangerous, destructive forces of nature that degrade air quality and
62 threaten human health and infrastructure (Larsen et al., 2018). However, fires are also a naturally
63 occurring disturbance needed to maintain the health and biodiversity of many ecosystems
64 (Goldammer et al., 2008; Weaver, 1974). In the Western United States, the natural fire cycle has
65 been completely disrupted and distorted by human interference (Fusco et al., 2016; Harvey,
66 2016), where large increases in fire size and occurrence are transpiring because of a buildup of
67 fuels caused by historically excessive fire suppression, increased settlement in the wildland
68 urban interface, and more favorable fire weather conditions (Abatzoglou & Williams, 2016;
69 Dennison et al., 2014; Mell et al., 2010; Stavros et al., 2014; Stephens & Ruth, 2005; Theobald
70 & Romme, 2007; Westerling et al., 2003). There is a need for balance between reducing the

71 hazards of wildland fires while maintaining forest health under the influence of a changing
72 climate.

73 Unless we can better understand and predict the deleterious impacts of wildland fire
74 smoke emissions on air quality and human health, it will be nearly impossible for society to
75 respond and adapt to this evolving and complex system. Informed land management policy that
76 utilizes prescribed fires to reduce fuel buildup and reinvigorate ecosystems in order to ultimately
77 minimize smoke exposure for downwind communities necessitates the ability to quantify the
78 composition, magnitude, and transport of smoke (Noss et al., 2006; Schweizer et al., 2018). In
79 the case of accidental or uncontrolled wildfires, the capability to accurately predict smoke
80 transport is necessary to alert sensitive populations and mitigate the overall impact of smoke on
81 human health (Larkin et al., 2009; McKenzie et al., 2006). Atmospheric models rely entirely on
82 geospatial databases of fire locations and estimated emissions (so-called fire emissions
83 inventories) to represent the contribution of fire emissions to downwind atmospheric
84 composition (Wiedinmyer et al., 2006). Two distinct approaches are traditionally used to create
85 these emissions inventories: a fuels-based bottom-up approach and a satellite-based top-down
86 approach (Seiler & Crutzen, 1980; Wooster et al., 2005).

87 The bottom-up approach calculates the mass of carbon emitted by a fire as the product of
88 burned area, fuel mass per unit area, the carbon fraction of fuel, and combustion completeness
89 (Seiler & Crutzen, 1980). This approach, also known as the carbon mass balance method,
90 operates under the explicit assumption that all burnt biomass carbon is volatilized and emitted to
91 the atmosphere (Ward & Radke, 1993). Although a fuels-based approach is the key feature of
92 bottom-up algorithms, most also rely on remote sensing observations from satellite sensors such
93 as the Moderate Resolution Imaging Spectroradiometer (MODIS) or Visible Infrared Imaging
94 Radiometer Suite (VIIRS) to determine burned area. Burned area can be calculated using active
95 fire detections, by assuming the entire landscape captured in the resolution of a single satellite
96 pixel burned, or burned area can be taken directly from higher level data products (Kaiser et al.,
97 2012; van der Werf et al., 2017; Wiedinmyer et al., 2011). Fuel mass per unit area is often
98 derived from either a biogeochemical model initialized with satellite observations and/or from
99 labor intensive fuel databases of fuel type and loading (McKenzie et al., 2012; Pettinari &
100 Chuvieco, 2016; Sandberg et al., 2001; van der Werf et al., 2017). Fuel carbon content is often
101 assumed based on laboratory measurements from previous studies or estimated using the sum of
102 CO₂, CO, and CH₄ emission factors (Akagi et al., 2011; McMeeking et al., 2009; Santín et al.,
103 2015; Susott et al., 1991; van der Werf et al., 2017; Yokelson et al., 1997). Combustion
104 completeness is calculated as a function of changes in visual landscape characteristics, reduction
105 in fuel moisture, increase in summer land surface temperature, tree cover, and/or daily fire
106 weather indices (Kaiser et al., 2012; van der Werf et al., 2017; Wiedinmyer et al., 2011).

107 The bottom-up approach requires an ecosystem-specific emission factor to convert total
108 carbon mass emissions to emissions of a particular trace gas or aerosol species (Akagi et al.,
109 2011; Andreae & Merlet, 2001). Emission factors are often attained from compilations of
110 previous studies categorized by fuel or vegetation type and show a wide range of natural
111 variability depending on the exact composition of fuel being burned and combustion conditions.
112 Certain species, including many volatile organic compounds (VOCs) and aerosols, rapidly
113 evolve in the atmosphere following emission, which necessitates emission factor estimates
114 derived only from measurements of young, fresh smoke. There are numerous particulate mass
115 (PM) emission factors published from ground and laboratory based studies, however in situ

116 airborne measurements of PM emission factors for Western US wildland fires are particularly
117 scarce (Akagi et al., 2011).

118 The top-down approach follows from Wooster et al. (2005), who showed that the burning
119 of dry vegetation yields the same amount of energy, regardless of fuel type. Top-down
120 inventories assume fire radiative power (FRP) observations from satellite remote sensing can be
121 used as a direct measurement of the amount of biomass consumed in a fire in an effort to bypass
122 the latency and uncertainty associated with variables required in bottom-up style inventories
123 (Ichoku & Kaufman, 2005). In the top-down approach, FRP is multiplied by a predetermined
124 coefficient, known as a smoke emission coefficient (C_c), to calculate fire emission rates of PM.
125 Smoke emission coefficients are constants derived for individual ecosystems by combining
126 multiple years of aerosol optical depth (AOD) remote sensing observations with a mass
127 extinction efficiency (MEE), a constant that relates particle extinction to particle mass (Giglio et
128 al., 2006; Ichoku et al., 2008; Kaiser et al., 2012). Ichoku et al. (2008) demonstrated that the
129 relationship between fire radiative energy (FRE), or temporally integrated FRP, and the emission
130 rate of PM could be quantified using AOD during a controlled laboratory-based experiment. The
131 smoke emission coefficient determined from the laboratory-based experiment agrees with
132 independent estimates derived from satellite measurements of FRP and AOD measured over
133 large-scale wildfires, which leads to the assumption that this approach can be extrapolated to
134 global scale observations of FRP and AOD.

135 There are dozens of top-down and bottom-up emissions inventories available for use in
136 atmospheric transport models. These inventories encompass wide ranges of spatial and temporal
137 scales and can be used to account for hundreds of individual pollutants emitted by fires
138 (Darmenov & da Silva, 2013; Ichoku & Ellison, 2014; Kaiser et al., 2009; Mota & Wooster,
139 2018; van der Werf et al., 2017; Wiedinmyer et al., 2011). The choice of which inventory to use
140 in modeling applications is crucial, because different fire emissions inventories can profoundly
141 disagree on the magnitude, composition, and temporal variability of fire emissions, especially
142 PM (Carter et al., 2020; Larkin et al., 2014; Liu et al., 2020; Pan et al., 2020). The underlying
143 cause of the disagreement is difficult to isolate, but could be an artifact of the various
144 assumptions used in each inventory. Most global emissions inventories are plagued with high
145 levels of uncertainty stemming from the individual datasets used to calculate emissions, which
146 further complicates the ability to isolate the cause of the discrepancies among inventories
147 (French et al., 2004; Urbanski et al., 2011; Wiedinmyer et al., 2011). For example, the detection
148 and quantification of active fire locations, FRP, and AOD using satellite remote sensing suffers
149 from the obscuration of the land surface by clouds or thick smoke, limited spatiotemporal
150 coverage or resolution, and instrument detection limits.

151 It is fundamentally challenging to correctly quantify biomass burning emissions due to
152 the highly variable composition and structure of the fuels that fires consume, and because fires
153 can rapidly change their behavior in response to dynamic meteorological or environmental
154 conditions (Kennedy et al., 2020; Liu, 2004; Schultz et al., 2008). The datasets used in global fire
155 emissions inventories attempt to capture these dynamics, but they often lack the spatial and
156 temporal resolution needed to fully encapsulate all of the individual components that influence
157 emissions. Intensive in situ measurements of smoke from the joint NASA/NOAA Fire Influence
158 on Regional to Global Environments and Air Quality (FIREX-AQ) campaign that was conducted
159 during the summer of 2019 provide a unique opportunity to evaluate the assumptions and
160 uncertainties in both top-down and bottom-up approaches for calculating fire emissions. During
161 FIREX-AQ, the NASA DC-8 aircraft was outfitted with a comprehensive instrument payload

162 that sampled smoke plumes from Western US wildland fires and Southeastern US prescribed and
163 agricultural fires. The plume sampling strategy for the western portion of the campaign consisted
164 of an above-plume, longitudinal run along the entire length of the plume to allow for nadir-
165 pointing remote sensing of the smoke followed by a set of plume transects perpendicular to the
166 direction of smoke transport where the aircraft sampled the plume in situ during a series of
167 sequentially-downwind, cross-sectional passes (Wiggins et al., 2020).

168 Measurements collected during FIREX-AQ provide the opportunity for a rare direct
169 comparison and evaluation of the traditional, lower resolution approaches to calculate fire
170 emissions at an unusually high spatial and temporal resolution. In this study, we calculate fire
171 total carbon and total PM emission rates from Western US wildland fires sampled during
172 FIREX-AQ using a novel, independent approach based on in situ smoke plume measurements.
173 Here, we integrate in situ trace gas and aerosol measurements with information on plume
174 thickness gleaned from airborne High-Spectral Resolution Lidar (HSRL) measurements to
175 calculate emission rates. Although this new approach is subject to its own uncertainties and
176 sources of error, we assume emission rate estimates derived from this approach are as close to
177 accurate as we can realistically achieve, because they are based on in situ measurements, and
178 their calculation doesn't require as many strong assumptions as the more traditional approaches.
179 We further capitalize on FIREX-AQ data to calculate fire emission rates using a high-resolution
180 top-down approach and a high-resolution bottom-up approach. The high-resolution top-down
181 approach (referred to as HSRL-GOES) uses airborne HSRL measurements of particle extinction
182 instead of satellite observations of AOD, and the high-resolution bottom-up approach (referred to
183 as Fuel2Fire) uses carbon emission estimates from the newly-developed Fuel2Fire carbon
184 emissions inventory that has been developed and optimized specifically to estimate emissions
185 from the fires sampled during FIREX-AQ. We also obtain emission rates from a traditional
186 bottom-up fire emissions inventory, Global Fire Emissions Database (GFED4.1s), and a
187 traditional top-down fire emissions inventory, Fire Energetics and Emissions Research
188 (FEERv1.0). GFED and FEER have much lower temporal and spatial resolutions (3-hr/daily,
189 0.25° and daily, 0.1° respectively) compared to the three high-resolution FIREX-AQ based
190 approaches. We evaluate the performance of GFED and FEER, along with the high-resolution
191 approaches, against the in situ measurement based approach to investigate potential bias and
192 assess the validity of the assumptions unique to each approach (Figure 1). We also investigate
193 and quantify uncertainty for all of the approaches used to calculate emission rates in this study.
194 The goal of this paper is to understand how the estimates of total carbon and PM emission rates
195 from traditional, lower resolution methods compare to the high-resolution estimates available for
196 the fires sampled during the FIREX-AQ campaign. The results of this analysis should be of keen
197 interest for the global wildfire emissions inventory community as well as atmospheric scientists
198 seeking to use airborne observations to constrain wildland fire aerosol emissions.

199 **2 Methods**

200 **2.1 Emission Rate Estimates from Global Inventories**

201 **2.1.1 GFED4.1s (Low-Resolution Bottom-up)**

202 GFED is a global fire emissions inventory that internally calculates carbon emission rates
203 using a traditional bottom-up approach as follows

$$E_C = BA \times FL \times CC \times F_C \quad (1)$$

where E_C is the carbon mass emission rate, BA is the burned area, FL is the fuel mass loading per area, CC is the combustion completeness (expressed as a percent), and F_C is the mass fraction of carbon in the fuel (van der Werf et al., 2017). GFED obtains burned area estimates from MODIS (MCD64A1), fuel loading and combustion completeness are derived from the Carnegie-Ames-Stanford Approach (CASA) biogeochemical model, and carbon mass fraction is defined per ecosystem from compilations of previous studies (Akagi et al., 2011; Andreae & Merlet, 2001; van der Werf et al., 2017).

To represent a traditional bottom-up approach, we use daily average carbon emission rates per area from GFED4.1s (<https://www.globalfiredata.org/>) to calculate daily average PM emission rates (E_{PM}) for the western fires sampled during FIREX-AQ as follows

$$E_{PM} = EF_{PM} \times \sum \frac{\hat{E}_C \times A_{P,GFED}}{F_C} \quad (2)$$

where EF_{PM} is the total particulate matter mass emission factor suggested by GFED for temperate forests (17.6 gPM kg-biomass consumed⁻¹), \hat{E}_C is the area-normalized daily carbon emissions in each GFED pixel, $A_{P,GFED}$ is the GFED pixel area (0.25° x 0.25°), and the summation is carried out over all GFED pixels within 0.25° of the centroid of the final United States Geological Survey Geospatial Multi-Agency Coordination (GeoMAC) fire perimeter for each fire. In Equation 2, we use the F_C suggested by GFED for temperate forests (0.489 kgC kg-biomass consumed⁻¹). The use of ecosystem level constant values for EF_{PM} and F_C is intended to provide good results in aggregate on the regional-to-global scales required by models, although individual fires will deviate from these specifications. GFED data is provided on a daily and a 3-hr basis in UTC time, and here we use the daily product. We convert from UTC time to local time by assuming daily emissions (local time) are equal to 75% of the emissions from the day a given fire was sampled by the DC-8 aircraft (local time) plus 25% of the emissions from the day after (local time).

We estimate relative uncertainty in E_C and E_{PM} estimates derived from GFED by propagating uncertainty through equation 1 and equation 2. For equation 1, we assume the following relative uncertainties: BA = 44%, FL = 111%, CC = 11%, and F_C = 10%. For equation 2, we assume EF_{PM} has a relative uncertainty of 36% and E_C has a relative uncertainty calculated by propagating uncertainty through equation 1. We obtain the relative uncertainty in the BA product used by GFED from an analysis of MODIS burned area by Giglio et al. (2018). FL and CC relative uncertainty are derived by taking the standard deviation divided by the average for all field measurements of Western US fuels as compiled by van Leeuwen et al. (2014) and updated by van der Werf et al. (2017). F_C relative uncertainty is defined as the standard deviation divided by the average in F_C values given by Akagi et al. (2011). The relative uncertainty in EF_{PM} is calculated as the standard deviation divided by the average of EF_{PM} derived from all previous studies of temperate forest EF_{PM} measurements used in GFED (Akagi et al., 2011;

241 Andreae and Merlet, 2001; van der Werf et al. 2017). The calculated relative uncertainty in
 242 GFED E_{PM} is 126% and E_C is 120% (Supplementary Table S1).

243 **2.1.2 FEERv1.0 (Low-Resolution Top-down)**

244 FEER is a global fire emissions inventory that calculates daily average E_{PM} using a
 245 traditional top-down approach as

$$246 \quad E_{PM} = C_e \times FRP \quad (3)$$

247 where C_e is an ecosystem-dependent predetermined smoke emission coefficient, and FRP
 248 observations are from MODIS. FEER derives C_e using multiple years of coupled MODIS AOD
 249 at 550nm and FRP observations and an assumed constant MEE at 550nm of $4.6 \text{ m}^2 \text{ g}^{-1}$ derived
 250 from previous studies (Reid et al., 2005b). Smoke emission coefficients have been predetermined
 251 by FEER and are provided globally at a $1^\circ \times 1^\circ$ resolution
 252 (<https://feer.gsfc.nasa.gov/projects/emissions/>) (Ichoku & Ellison, 2014).

253 Daily average E_{PM} estimates are provided at a $0.1^\circ \times 0.1^\circ$ resolution through a coupling
 254 of FEER smoke emission coefficients and MODIS FRP observations (FEERv1.0-G1.2). For the
 255 traditional top-down approach, we calculate daily average E_{PM} estimates for each of the western
 256 fires sampled during FIREX-AQ as

$$257 \quad E_{PM} = \sum \hat{E}_{PM_{FEER}} \times A_{P,FEER} \quad (4)$$

258 where $\hat{E}_{PM_{FEER}}$ is the area-normalized daily average E_{PM} from each FEER pixel, and $A_{P,FEER}$ is
 259 the FEER grid cell area ($0.1^\circ \times 0.1^\circ$). The summation is over all FEER grid cells per fire. FEER
 260 grid cells are included if they are within 0.1° of the centroid of the final GeoMAC fire perimeter
 261 for each fire. We convert from UTC time to local time following the same approach described in
 262 section 2.1.1. We also calculate the average FEER C_e for the fires sampled during the western
 263 portion of the FIREX-AQ campaign as the average of all C_e estimates in every 1° grid cell that
 264 encompassed at least a fraction of the final GeoMAC perimeter of a fire.

265 We estimate relative uncertainty in FEER E_{PM} estimates by propagating uncertainty
 266 through equation 3. We calculate the relative uncertainty of C_e as the standard deviation divided
 267 by the mean of all extracted FEER C_e values used in this analysis, and we obtain the relative
 268 uncertainty of MODIS FRP from Freeborn et al. (2014). FEER C_e for Western US wildland fires
 269 has a relative uncertainty of 73% and FRP has a relative uncertainty of 27%, yielding a relative
 270 uncertainty in FEER E_{PM} of 78% (Supplementary Table S1).

271 **2.2 Emission Rate Estimates from FIREX-AQ**

272 **2.2.1 In Situ Measurement Approach (High-Resolution)**

273 We capitalize on the intensive, high spatial and temporal resolution smoke plume
 274 measurements from the DC-8 aircraft during FIREX-AQ to calculate E_C and E_{PM} via a novel in
 275 situ measurement-driven approach. We assume fire emission rates over time are equal to the flux
 276 of smoke as it passes through a vertical slice of the smoke plume, represented as an HSRL

277 curtain measured during in situ transects (Figure 1). We calculate E_C and E_{PM} for each wildland
 278 fire sampled during FIREX-AQ on a sub-plume (per transect) basis as

$$279 \quad E_X = \overline{WS} \times \overline{GS} \times \sum_{t_{start}}^{t_{end}} \Delta X_t \times H_t \Delta t \quad (5)$$

280 where E_X is the emission rate of species X (either carbon or PM), \overline{WS} is the transect average wind
 281 speed, \overline{GS} is the transect average ground speed, ΔX_t is the excess concentration of species X
 282 averaged over 10 second intervals to match the horizontal resolution of the HSRL data collected
 283 at aircraft measurement time t , and H_t (m) is the plume thickness measured by the nadir and
 284 zenith pointing HSRL profiles at aircraft measurement time t (Hair et al., 2008). This approach
 285 assumes that the vertical distribution of each species is uniform, and the lidar is used to find the
 286 vertical extent of the plume. Excess concentrations are calculated by subtracting a background
 287 concentration defined as the average concentration 5-10 seconds prior to the start of the transect
 288 and 5-10 seconds after the end of the transect. In a few cases, the PM background is elevated
 289 during the time interval used to define a background such that the excess mixing ratio is
 290 computed as a negative value, and these cases are excluded from the analysis. The time interval
 291 from t_{start} to t_{end} is equal to the length of time to complete each transect, and Δt is ~ 10 seconds,
 292 which is the horizontal resolution of the HSRL data. H_t is calculated as the sum of HSRL profile
 293 bin heights (Δz_t) where the particle backscatter coefficient (β_t) is greater than $1 \text{ km}^{-1} \text{ sr}^{-1}$, which
 294 was larger than the average background scattering and defined the smoke plume edges for the
 295 cases sampled.

$$296 \quad H_t = \sum_{z=0}^{z=\infty} \Delta z_t [\beta_t > 1 \text{ km}^{-1} \text{ sr}^{-1}] \quad (6)$$

297 In exceptionally dense smoke plumes, the HSRL laser light was fully attenuated before it
 298 could completely pass through the smoke plume edge, and for these cases we assume the smoke
 299 plume extended to the surface and neglect the missing portion of the plume above the aircraft.
 300 This approach is reasonable as the aircraft tended to sample the smoke plumes near the top of the
 301 atmospheric boundary layer, which places a weak upper constraint on the top of the plume just as
 302 the surface places a lower constraint on boundary layer mixing processes. An alternative
 303 approach to calculate plume thickness using HSRL observations leverages the ratio of the
 304 backscatter coefficient in a single HSRL bin to the sum of all backscatter coefficients in a
 305 vertical column. We estimate the sensitivity of plume thickness to these two approaches and
 306 discover strong agreement (slope = 0.72, $r = 0.83$), although the alternative approach estimates
 307 slightly lower plume thickness on average (Figure S1). We ultimately choose the approach to
 308 calculate plume thickness as outlined in equation 6 in an effort to avoid additional uncertainty
 309 from relying more heavily on the backscatter coefficient, which may be confounded by changes
 310 in aerosol size and/or optical properties rather than mass loading.

311 Total PM is calculated as the sum of organic aerosol (OA), sulfate, nitrate, ammonium,
 312 and black carbon aerosol (BC) reported at standard temperature and pressure conditions and
 313 converted to ambient volumetric units. The 50% geometric transmission diameter for the AMS is
 314 approximately 600nm, which sufficiently captures the size range for the majority of biomass
 315 burning derived particles, with the exception of supermicron ash particles (Adachi et al., 2021;
 316 Moore et al., 2021). All components of the submicron non-refractory total PM concentrations are
 317 measured using an Aerodyne Time of Flight Aerosol Mass Spectrometer (ToF-AMS)
 318 (Canagaratna et al., 2007; DeCarlo et al., 2006; Guo et al., 2021). Refractory BC mass
 319 concentrations are provided by a Single Particle Soot Photometer (SP2, Droplet Measurement
 320 Technologies). Total carbon is calculated as the sum of CO_2 , CO , CH_4 , organic carbonaceous

321 aerosol (OC), and BC aerosol. OC is estimated using the OA to OC ratio provided by the ToF-
 322 AMS. The CO₂ mixing ratio measurements are obtained using a non-dispersive infrared (IR)
 323 spectrometer (LICOR, Inc. Model 7000) adapted for aircraft measurements in a method similar
 324 to Vay et al. (2003), while CO and CH₄ mixing ratios are obtained from mid-IR laser absorption
 325 spectrometry (Sachse et al., 1991). All three trace gas species were calibrated in-flight with
 326 standards from the National Oceanic and Atmospheric Administration Earth Science Research
 327 Laboratories (NOAA ESRL) traceable to World Meteorological Organization (WMO) scales.
 328 The trace gas measurements were converted from mole fractions to ambient volumetric units by
 329 multiplying the mixing ratio by the ratio of the molecular weight to the molecular volume at
 330 ambient temperature and pressure conditions.

331 We estimate relative uncertainty in E_C and E_{PM} using equations 5 and 6. We calculate the
 332 following relative uncertainties: WS = 20%, GS = 3%, H_t = 28%, ΔC = 56%, and ΔPM = 67%.
 333 The relative uncertainty for each variable is assumed to be equal to the mean divided by the
 334 standard deviation of observations collected during all smoke plume transects. The computed
 335 relative uncertainty in E_C is 66% and the relative uncertainty in E_{PM} is 75% (Supplementary
 336 Table S1).

337 **2.2.2 Fuel2Fire (High-Resolution Bottom-up)**

338 E_C estimates for all FIREX-AQ wildland fires derived using a bottom-up style approach
 339 are publicly available on the FIREX-AQ data archive
 340 (<https://doi.org/10.5067/SUBORBITAL/FIREXAQ2019/DATA001>). This dataset, the Fuel2Fire
 341 carbon emissions inventory, is optimized and designed to estimate carbon emissions specifically
 342 for the fires sampled during FIREX-AQ. We use Fuel2Fire E_C estimates for the high-resolution
 343 bottom-up approach to estimate E_C and E_{PM} on a per transect basis for each of the fires included
 344 in this analysis. As a bottom-up inventory, Fuel2Fire calculates E_C in the same way as GFED,
 345 following equation 1. The Fuel2Fire emissions inventory derives burned area using a
 346 combination of active fire detections from MODIS, VIIRS, and/or Geostationary Operational
 347 Environment Satellite Program (GOES-16 and 17 ABI L2 +). Active fire pixels from one or
 348 more of these active fire detection products are selected to best match ground-verified
 349 interagency situational reports from fire management teams, as well as GeoMAC fire perimeters.
 350 Fuel2Fire determines fuel loading using ultra high-resolution (30 meter) fuels data from the
 351 Fuels Characteristics and Classification System (FCCS) and models combustion completeness as
 352 a function of daily fire weather danger ratings. Total daily carbon emissions are temporally
 353 distributed using a diurnal cycle of fire activity derived from geostationary satellite observations
 354 of FRP from GOES-16 and 17. Fuel2Fire assumes F_C is 0.5 kg kg⁻¹ (McMeeking et al., 2009;
 355 Santín et al., 2015; Yokelson et al., 1997). The archived carbon emissions data has a native
 356 temporal resolution that matches GOES-16 and 17 data (5 minutes) and is linearly interpolated to
 357 1 Hz data for consistency with the aircraft data. E_C estimates from Fuel2Fire extend over the
 358 course of an entire 24-hour day (local time) that a given fire was sampled during FIREX-AQ.

359 We convert E_C estimates from the Fuel2Fire inventory to E_{PM} as follows

$$360 \quad E_{PM} = \frac{E_C \times EF_{PM}}{F_C} \quad (7)$$

361 Here, we obtain E_C from the Fuel2Fire inventory, while EF_{PM} is calculated using aircraft
 362 observations. We choose to calculate EF_{PM} from in situ observations as opposed to assuming
 363 EF_{PM} from a compilation of previous studies in order to investigate the potential influence of the
 364 choice in EF_{PM} on differences in emission rate estimates. F_C is assumed to be 0.5 kg C kg⁻¹ fuel⁻¹,

365 but we note F_C can vary from 0.35-0.55 (Akagi et al., 2011; Burling et al., 2010; McMeeking et
 366 al. 2009; Susott, 1996). We calculate EF_{PM} for each in situ smoke plume transect using airborne
 367 measurements following the carbon mass balance approach (Ward & Radke, 1993; Yokelson et
 368 al., 1996, 1999). Although the smoke age and, thus, probability of plume processing increases as
 369 a function of downwind distance from the fire, we assume PM is conserved over the relatively
 370 short period of time (0.5-7 hrs) that the smoke has been exposed to atmospheric aging processes
 371 when it was sampled by the DC-8 and attribute any changes in mass concentration to variability
 372 in fire activity (Garofalo et a., 2019; Hodshire et al., 2019). The time of emission is not the same
 373 as when the DC-8 sampled the plume, so we correct for this time offset by adding smoke age to
 374 the time of emission when determining the Fuel2Fire total carbon emission rates on a sub-plume,
 375 per transect basis. The smoke age is calculated for each point on the DC-8 transect assuming
 376 horizontal straight line advection of the smoke plume at the DC-8 measured wind speed
 377 (Wiggins et al., 2020).

378 We estimate relative uncertainty in E_C and E_{PM} derived from Fuel2Fire by propagating
 379 uncertainty through equation 7. The relative uncertainty in E_C is assumed to be 55%, calculated
 380 by taking the average divided by the standard deviation of all computed E_C estimates for each
 381 fire and every transect included in this analysis. The relative uncertainty in EF_{PM} is 39%,
 382 computed as the mean of all calculated EF_{PM} for all fires divided by the standard deviation. The
 383 relative uncertainty in E_{PM} is thus 67% (Supplementary Table S1).

384 2.2.3 HSRL-GOES (High-Resolution Top-down)

385 We use FIREX-AQ aircraft-based HSRL measurements of aerosol extinction and
 386 geostationary satellite observations of FRP from GOES to calculate E_{PM} using a high-resolution
 387 top-down approach, referred to as HSRL-GOES. We use the same equation that is used in FEER
 388 (Equation 3) to calculate E_{PM} for the western fires sampled during FIREX-AQ on a per transect
 389 basis for the high-resolution top-down approach. Instead of using MODIS FRP, we obtain FRP
 390 from the GOES-16 and GOES-17 ABI L2 + Fire/hot spot Detection and Characterization product
 391 from the Wildfire Automated Biomass Burning Algorithm processing system (Schmidt, 2019).
 392 GOES has an exceptionally high temporal resolution (~5-15 mins) with FRP observations that
 393 cover the entire continental US at a spatial resolution of 2km (Schmidt, 2019). We time align
 394 GOES FRP observations to match the in situ plume sampling time by adding the smoke age to
 395 the FRP observation time, and we include all FRP observations within 4km (the spatial
 396 resolution of GOES-16 and 17) of a given fire's final GeoMAC perimeter centroid. FRP per
 397 transect is calculated as the sum of all instantaneous FRP observations for a given fire averaged
 398 over the in situ plume sampling time for a given transect. The smoke emission coefficient is also
 399 calculated for each fire on a per transect basis as follows:

400

$$401 \quad C_e = \frac{\overline{WS \times GS}}{\overline{MEE \times FRP_f}} \times \sum_{t_{start}}^{t_{end}} \Delta AOT_t \Delta t \quad (8)$$

402

403 where $\overline{FRP_f}$ is the time-aligned, transect-average GOES FRP, \overline{MEE} is the transect average MEE
 404 calculated from in situ measurements as described below, and ΔAOT is aerosol optical thickness

405 derived from vertically integrating the background-subtracted 532nm HSRL particle extinction
 406 coefficient ($\Delta\alpha$) as described by

407

$$408 \quad \Delta AOT_t = \int \Delta\alpha_t \Delta z \quad (9)$$

409

410 HSRL is not able to collect measurements immediately above and below the aircraft. We linearly
 411 interpolated through the 60m aircraft gap in the HSRL curtains to account for the missing data.
 412 Background extinction is defined as the average HSRL extinction profile 10 sec before and after
 413 the smoke plume transect. In cases where the laser light fully attenuated before it reached the
 414 bottom of the plume, we assume the plume extended to the surface and extrapolate extinction to
 415 the ground using the closest measurement to the surface. In limited cases where the beam is
 416 completely attenuated in the zenith direction, we integrate over the measured range but do not add
 417 any correction as this is expected to be a relatively small contribution as the aircraft was typically
 418 flying near the top of the atmospheric boundary layer near plume top. We use the high-resolution
 419 in situ measurements from the DC-8 to calculate MEE; however, we note that most top-down
 420 inventories (such as FEER) assume a constant MEE of $4.6 \text{ m}^2 \text{ g}^{-1}$ derived from previous studies.
 421 We calculate transect average MEE as the slope of a reduced major axis regression with a
 422 forced zero intercept between total PM and the dry aerosol extinction coefficient at 532nm for
 423 each transect. The extinction coefficient is calculated as the sum of dry scattering and absorption
 424 coefficients measured by a TSI-3563 Nephelometer at 550nm and a 3-wavelength Particle Soot
 425 Absorption Photometer at 532nm (PSAP, Radiance Research) respectively. Scattering
 426 coefficients are converted to 532nm to match the absorption coefficients using the angstrom
 427 exponent as calculated by the blue and green channels from the nephelometer. Scattering
 428 coefficients are corrected for truncation errors following Anderson and Ogren (1998), and PSAP
 429 absorption data are corrected following Virkkula et al. (2010). The aerosol extinction
 430 humidification factor, $f(\text{RH})$ is assumed to be unity, which is consistent with the FIREX-AQ in-
 431 plume measurements.

432 We estimate the uncertainty in HSRL-GOES E_{PM} by propagating uncertainty through
 433 equation 3, where the relative uncertainty in C_e derived following equation 8 is calculated as the
 434 mean C_e from all fires divided by the standard deviation (67%), and the relative uncertainty in
 435 FRP is assumed to be 40% (Li et al., 2020). The relative uncertainty in HSRL-GOES E_{PM} is thus
 436 77% (Supplementary Table S1).

437 **2.3 Comparison of Approaches**

438 We summarize the approaches and relevant equations used in this study to calculate E_c
 439 and E_{PM} in Table 1. We evaluate emission rate estimates between the high-resolution bottom-up
 440 (Fuel2Fire) and top-down (HSRL-GOES) based approaches against the in situ approach on a per
 441 transect basis for individual wildland fires sampled during FIREX-AQ. The relationship between
 442 the different approaches is quantified using the slope of a reduced major axis regression with a
 443 forced zero intercept, a Pearson's correlation coefficient, and root mean square error (RMSE).
 444 These calculations are performed as a campaign level summary that includes all transects and all
 445 fires and for each fire individually.

446 We compare daily (24 hour local time) average E_{PM} estimates from the three high
 447 resolution approaches (In Situ, Fuel2Fire, and HSRL-GOES) to daily average estimates derived

448 from lower resolution global fire emissions inventories (GFED and FEER). These comparisons
449 are performed on daily average emission rate estimates, as opposed to estimates on a per transect
450 basis, because of the lower temporal resolutions of GFED (3 hr/daily) and FEER (daily).

451 **2.4 Smoke Emission Coefficients**

452 The high spatial and temporal resolution of the in situ, bottom-up (Fuel2Fire), and top-
453 down (HSRL-GOES) based approaches provide the opportunity to evaluate smoke emission
454 coefficients that are usually derived using many years of data. Smoke emission coefficients for
455 PM are calculated as the slope of a reduced major axis regression with a forced zero intercept
456 between GOES FRP time aligned to the transect sampling time versus E_{PM} for each of the three
457 high resolution approaches. These computations are also executed as a campaign level summary
458 and for each fire individually. We compare our C_e from the high resolution approaches to the
459 average FEER C_e for the western fires sampled during FIREX-AQ.

460 **3 Results and Discussion**

461 **3.1 Total Carbon Emission Rates**

462 The derivation of the variables used to calculate E_C using bottom-up approaches are
463 based on assumptions that can lead to both under and overestimation, depending on the data
464 products leveraged by a given fire emissions inventory. We uncover a significant relationship
465 between E_C per transect derived from the high-resolution bottom-up approach (Fuel2Fire) and
466 the in situ approach as shown in Figure 2 (slope = 1.00, $r = 0.82$). However, there is also a non-
467 trivial level of scatter in this relationship (RMSE = 67%), and individual fires considered
468 separately have different correlations and regression slopes.

469 From Figure 3, we similarly find strong, linear correlations between daily fire average E_C
470 from the in situ measurement based estimates and Fuel2Fire (slope = 1.09, $r = 0.92$, RMSE =
471 61%) and GFED (slope = 0.20, $r = 0.87$, RMSE = 132%). The daily average E_C estimated using
472 Fuel2Fire are marginally higher than estimates derived from the in situ approach, while the
473 GFED estimates are 80% lower. The strong correlation, but significant offset between GFED and
474 the in situ measurement based approach implies that there may be a systematic bias in one or
475 more of the variables used to calculate the mass of biomass consumed in some traditional
476 bottom-up inventories. In this section, we examine the assumptions and uncertainty in individual
477 variables used to calculate E_C using a bottom-up approach in an effort to understand the
478 differences in E_C estimates derived from Fuel2Fire and GFED relative to the in situ approach.

479 **3.1.1 Carbon Mass Balance**

480 The key assumption in a bottom-up approach is that all burnt carbon is volatilized and
481 released into the atmosphere. This carbon mass balance assumption has recently been
482 scrutinized, because not all fuel that has been thermally altered by a fire is emitted to the
483 atmosphere (Santín et al., 2015; Surawski et al., 2016). Some of the burnt fuel remains on the
484 ground as charred biomass. If the carbon mass balance assumption does not hold, then this could
485 potentially cause an overestimation of carbon emissions derived from bottom-up approaches by
486 up to 50% in temperate forests, depending on levels of combustion completeness (Santín et al.,
487 2015). Our results do not show significant evidence of bias in E_C estimates from Fuel2Fire, but
488 do show a distinct low bias in estimates from GFED. This suggests there are underlying
489 confounding factors to disentangle before it is possible to determine if the assumptions inherent

490 in the carbon mass balance approach are responsible for a significant bias in bottom-up
491 inventories.

492 **3.1.2 Burned Area**

493 The two methods for calculating burned area using a bottom-up approach operate under
494 specific assumptions that could cause either an over or under estimation of carbon emissions.
495 The active fire based method has the potential to overestimate burned area, because it assumes
496 all the area within the resolution of a single active fire detection is burned. Conversely, the
497 burned area based method using MODIS burned area data products (MCD65A1) has been shown
498 to underestimate burned area, because of high omission error in grid cells with smaller
499 proportions of burned area (Boschetti et al., 2019). A recent validation study that compares 500-
500 meter resolution MODIS burned area products (MCD64A1) against 30-meter resolution Landsat
501 data found MODIS underestimated global burned area by 54% (Boschetti et al., 2019). However,
502 MODIS burned area products also have a non-trivial level of uncertainty, approximately 44%
503 (Giglio et al., 2018).

504 Fuel2Fire, calculates burned area using the active fire approach, while GFED uses
505 MODIS burned area data products. GFED4.1s attempts to address the known small fire driven
506 burned area underestimation from MODIS using a supplementary algorithm known as the small
507 fire boost (Randerson et al., 2012; van der Werf et al., 2017). We compare the GFED and
508 Fuel2Fire burned area estimates in Figure S2, which are in good agreement for the western fires
509 sampled during FIREX-AQ (slope = 0.97, $r = 0.93$). This indicates that the differences in the two
510 approaches to calculate burned area are not responsible for the low bias we see in GFED
511 emission rate estimates.

512 **3.1.3 Combustion Completeness**

513 All state-of-the-art approaches to calculate combustion completeness rely on daily or
514 monthly average observations, and therefore cannot accurately estimate the pronounced sub-
515 daily changes in combustion completeness that occur throughout the diurnal cycle of fire
516 activity. Instead, these methods assume combustion completeness can be estimated using
517 observations averaged over large areas. Combustion completeness in the Fuel2Fire inventory is
518 based on daily fire weather indices, where higher levels of fire danger equate to higher
519 consumption rates, while GFED relies on a biogeochemical model with a monthly time step to
520 estimate combustion completeness.

521 We expect GFED combustion completeness to be overestimated on days with low fire
522 activity and underestimated on days with high fire activity as a result of the monthly averaging
523 scheme, however that is not the observed trend (Figure 3). Instead, GFED underestimates E_C for
524 almost every fire included in this analysis. It is therefore unlikely that differences in the
525 approaches to calculate combustion completeness strongly contribute to the systematic low bias
526 found in GFED E_C estimates.

527 **3.1.4 Fuel Loading**

528 The complexity and variability of fuel type (or land cover) and loading are difficult to
529 accurately represent and validate. Labor intensive high spatial resolution fuel databases, such as
530 the FCCS database used in the Fuel2Fire inventory, are derived from a compilation of previous
531 remote sensing studies, government databases, photos, in situ measurements, and expert opinion
532 (Ottmar et al., 2007). The spatial resolution of FCCS is 30m; however, this resolution is achieved

533 through the extrapolation of field-based measurements to ecosystem scales, which relies on a
534 number of strong assumptions that infer the distribution and composition of fuels from the same,
535 similarly-aged, spectrally-similar ecosystems are roughly spatially constant. Fuels are constantly
536 changing in response to seasonal, environmental, and anthropogenic forcing, but the laborious
537 effort required to develop fuel databases severely restricts the rate at which they can be updated.
538 As a result, fuel bed databases can remain unchanged and out of date for a number of years
539 before updates are implemented. This delay can exacerbate the uncertainty and error in fuel
540 loading estimates. Model based estimates of fuel loading that rely on remote sensing
541 observations of surface characteristics, like those used in GFED, are similarly challenged by the
542 limited number of field measurements available to validate estimates. Potential bias stemming
543 from fuel loading estimates can be negative or positive, depending on the accuracy of initial
544 estimates and if the database or model correctly implements changes in fuel loading following
545 ecosystem disturbance mechanisms including fire.

546 We discover an exceptional uncertainty in GFED fuel loading (111%), however the
547 uncertainty in the FCCS fuels database used by Fuel2Fire is estimated to be much lower (~70%)
548 (Keane et al., 2013) (Supplementary Table S1). Combined with the lack of evidence that burned
549 area or combustion completeness significantly contribute to GFED emission estimate bias, this
550 implies the differences in fuel loading estimates from the model used in GFED versus the high-
551 resolution fuels database (FCCS) used in Fuel2Fire is the most likely culprit for the persistent
552 underestimation of GFED E_C estimates from Western US wildland fires. The agreement we see
553 in E_C estimates from Fuel2Fire versus the in situ based approach provides confidence for the use
554 of high resolution fuels databases such as FCCS (Figure 2).

555 Previous studies aimed at quantifying uncertainty in the parameters used by bottom-up
556 inventories to calculate emissions have also identified fuel loading as a major source of
557 uncertainty (French et al., 2004; Urbanski et al., 2011; Prichard et al., 2019). Furthermore, fuel
558 loading uncertainty likely fluctuates considerably as a function of vegetation type, due to scarce
559 field validation studies for certain ecosystems and/or mapping errors. Our results highlight the
560 need for additional field validation studies to constrain fuel loading estimates.

561 **3.2 Total PM Emission Rates**

562 We find the strong relationship between Fuel2Fire and the in situ based method persists
563 for E_{PM} at a sub-plume scale, albeit with a similarly high level of scatter as shown in Figure 4a
564 (slope = 0.90, $r = 0.77$, RMSE = 61%). We derive E_{PM} from the in situ FIREX-AQ
565 measurements on a per transect basis in order to minimize the potential influence of emission
566 factor uncertainty in Fuel2Fire E_{PM} estimates. The high level of spread in the data is likely an
567 artifact of the uncertainty in E_C from Fuel2Fire caused by the biases and sources of uncertainty
568 discussed in Section 3.1, most notably the impacts of fuel loading uncertainties. Additionally,
569 this comparison is based on the assumption that the transport of fire emissions from the ground
570 to the in-situ transect is accurately modeled in both space and time.

571 Figure 4b shows a significant relationship between E_{PM} calculated using HSRL-GOES
572 and the in situ approach at a sub-plume scale (slope = 1.04, $r = 0.82$, RMSE = 48%). While there
573 is a marginally lower level of scatter in this relationship as shown by the RMSE, HSRL-GOES
574 slightly overestimates E_{PM} on the lower end of the scale compared to the in situ approach. This
575 overestimate implies from Equation 3 that either or both the GOES FRP and C_e for these fire
576 transects are biased high, where it follows from Equation 12 that the latter may be influenced by
577 a low estimate of the smoke MEE or a high estimate of the optical thickness. A high optical

578 thickness bias might be due to the extrapolation of HSRL extinction to the surface for cases
579 when the laser light fully attenuates; although, we note the bias is most significant for the lower
580 emission rates, which might discount this hypothesis.

581 We find strong correlations between daily average E_{PM} estimates from the in situ
582 approach versus estimates from both of the high-resolution approaches, Fuel2Fire (slope = 1.04,
583 $r = 0.93$, RMSE = 39%) and HSRL-GOES (slope = 1.18, $r = 0.89$, RMSE = 47%) (Figure 5). The
584 correlation is weaker and the spread is larger between the in situ based estimates and estimates
585 from the lower-resolution global inventories, GFED and FEER. The systematic low bias seen in
586 GFED daily average E_C estimates is also seen for daily average E_{PM} for all but the smallest fires
587 (slope = 0.21, $r = 0.85$, RMSE = 104%). FEER slightly underestimates E_{PM} from larger fires that
588 emit relatively more PM and overestimates E_{PM} from smaller fires that emit relatively less PM
589 (slope = 1.38, $r = 0.64$, RMSE = 55%). FEER provides no E_{PM} estimates for the Castle fire on
590 both days of sampling, and we exclude these zero estimates from this fire when computing the
591 linear regression and correlation coefficient given their disproportionate weight in skewing the
592 regression.

593 Global fire emissions inventories are known to significantly differ on E_{PM} estimates from
594 temperate fires, especially in North America (Nikonovas et al., 2017; Pan et al., 2020). In the
595 following sections we use the high resolution airborne in situ measurements of smoke plumes
596 collected during FIREX-AQ to isolate the assumptions and variables responsible for the
597 discrepancy and quantify their relative contributions.

598 3.2.1 Emission Factors

599 Emission factors are used in bottom-up approaches to convert carbon emissions to
600 emissions of a specific trace gas or particulate species, and emission factor estimates usually
601 come from compilation studies that include in situ measurements from wildland fires and
602 laboratory experiments (Akagi et al., 2011; Andreae, 2019; Andreae & Merlet, 2001; May et al.,
603 2014). The use of such emission factors relies on the assumption that the most representative
604 value can be approximated as the mean of all previous studies. In reality, emission factors are
605 dynamic and vary as a function of combustion completeness, which can fluctuate both spatially
606 and temporally for a given fire. Laboratory studies struggle to represent the complexity of a
607 wildland fire and can disagree with in situ measurements, while in situ measurements are subject
608 to sampling bias (Hodshire et al., 2019; Yokelson et al., 2013). For example, airborne based
609 measurements tend to be limited to daytime sampling of well-developed plumes that have risen
610 to an altitude that is accessible by the aircraft. Consequently, these measurements may be biased
611 towards flaming combustion because nighttime and/or smoldering emissions resulting from less
612 energetic fire activity are not being sampled (Burling et al., 2011; Prichard et al., 2020; Wiggins
613 et al., 2021). The suggested EF_{PM} for temperate forests from GFED is 17.6 g kg^{-1} , and the mean
614 EF_{PM} we calculated using FIREX-AQ in situ airborne measurements is $15.8 \pm 4.3 \text{ g kg}^{-1}$, which
615 is well within range of the suggested EF_{PM} from GFED. Our results suggest EF_{PM} does not
616 strongly contribute to bias in bottom-up emission rate estimates from Fuel2Fire or GFED for the
617 fires sampled during FIREX-AQ. However, we acknowledge this analysis focused exclusively
618 on fires with well-developed plumes that were sampled during the daytime, and thus may not be

619 subject to E_{PM} discrepancies that can occur as a result of under sampled smoldering
620 combustion.

621 **3.2.2 Smoke Emission Coefficient (C_e)**

622 Smoke emission coefficients used by top-down inventories to convert FRP to E_{PM} are
623 typically derived using multiple years of AOD and FRP observations, but here we use high
624 resolution measurements from FIREX-AQ to calculate C_e over a limited duration for a small
625 number of fires. We find strong to moderate linear relationships between GOES FRP
626 observations and the calculated emission rates from the high resolution in situ approach ($C_e = 5.0$
627 gPM MW^{-1} , $r = 0.75$, $\text{RMSE} = 165\%$), Fuel2Fire ($C_e = 8.2 \text{ gPM MW}^{-1}$, $r = 0.94$, $\text{RMSE} = 46\%$),
628 and HSRL-GOES ($C_e = 8.4 \text{ gPM MW}^{-1}$, $r = 0.72$, $\text{RMSE} = 75\%$) (Figure 6). Individual fires
629 have significantly different C_e , and vary depending on which approach was used to calculate E_{PM}
630 (Table 3), which highlights the sensitivity and natural variability of this parameter. All three of
631 the high resolution approaches estimate a lower C_e for the set of Western US wildland fires
632 included in this study compared to the estimated C_e from FEER (10.6 gPM MW^{-1}). However, the
633 calculated C_e are within the large uncertainty (50%) of the C_e for western US fires derived from
634 FEER, and the RMSE is substantial for the in situ approach and HSRL-GOES.

635 Fuel2Fire temporally distributes emissions using the diurnal cycle of GOES FRP
636 observations, which explains the exceptionally strong linearity and correlation between GOES
637 FRP and E_{PM} estimates in Figure 6b. HSRL-GOES E_{PM} estimates shown in Figure 6c continue to
638 have a slight high bias on the lower end of the scale. We find a high bias in E_{PM} for the Castle
639 fire in all three high resolution approaches compared to what would be expected based on the
640 overall campaign level relationship between FRP and emission rates (Figure 6). The Castle fire
641 had the lowest average excess PM concentrations per transect out of all the fires included in this
642 analysis. The elevated emission rates from all three approaches indicate GOES likely missed
643 some of the FRP, likely due to low temperature smoldering or cloud cover, which is consistent
644 with the low fire severity measured in post-burn satellite data.

645 FEER uses MODIS FRP observations to calculate C_e , but we use GOES FRP. There
646 could be a potential offset between FRP observations between MODIS and GOES as a result of
647 differences in instrument resolution and saturation levels as well as overpass time effects (Li et
648 al., 2019; Xu et al., 2021). The coarse spatial resolution of GOES (2km) limits its capability to
649 detect cool or small fires with low FRP, and could result in an underestimation of FRP by up to
650 50% globally (Freeborn et al., 2008), which would explain the difference in C_e estimated using
651 the high resolution approaches versus FEER. We use GOES FRP because of the exceptionally
652 high time resolution (5-15 mins) over the continental US versus the twice daily temporal
653 resolution of MODIS or VIIRS. This allows for a more direct comparison between in situ
654 measurements and remote sensing observations.

655 Previous studies have suggested E_{PM} and thus C_e calculated using MODIS AOD may be
656 systematically biased low, because of a discrepancy between observed AOD from MODIS
657 versus AERONET and MISR (Pan et al., 2020). However, we find agreement between E_{PM}
658 calculated independently of MODIS AOD and E_{PM} estimated from FEER, which relies on
659 MODIS AOD observations. The results suggest MODIS AOD observations can be used to

660 accurately represent atmospheric aerosol mass loading of particulates emitted by Western US
661 wildland fires.

662 **3.2.2.1 Mass Extinction Efficiency (MEE)**

663 The conversion of FRP to PM assumes that variability in particle extinction, and thus
664 AOD, is driven by changes in aerosol mass concentration rather than aerosol intensive
665 properties. Estimates of particle mass extinction efficiency (MEE) are essential to the conversion
666 of AOD to total PM. However, aerosol extinction and other optical properties depend on particle
667 size, morphology, and chemical composition (Seinfeld & Pandis, 2006). The characteristics of
668 biomass burning aerosols are known to vary with fuel type and combustion completeness
669 (McClure et al., 2020; Reid et al., 2005a; Reid et al., 2005b). Furthermore, the physical and
670 optical properties of smoke aerosols rapidly evolve following emission as a result of
671 photochemical aging and aerosol microphysical processes (Akagi et al., 2012; Cappa et al., 2020;
672 Garofalo et al., 2019; Hodshire et al., 2019; May et al., 2014; Shingler et al., 2016). Particle
673 evolution via these processes is additionally influenced by external factors, such as the fire size,
674 rate of dilution, and background aerosol concentrations (Hodshire et al., 2019). The assumption
675 that variability in AOD is entirely due to changes in particle concentration oversimplifies the
676 complex interactions of smoke particle microphysical processes and photochemical aging. Some
677 top-down inventories attempt to reconcile this discrepancy by calculating a separate C_e for each
678 individual ecosystem. However, this is likely not sufficient to fully address the variability in
679 smoke aerosol extinction that often occurs even in smoke plumes from fires within a single
680 ecosystem type.

681 Top-down inventories, including FEER, usually assume a constant MEE of $4.6 \text{ m}^2 \text{ g}^{-1}$
682 based on a compilation of previous studies (Ichoku & Kaufman, 2005; Reid et al., 2005b). The
683 compilation found MEE varied between $3.4 - 5.1 \text{ m}^2 \text{ g}^{-1}$ for biomass burning particles of all ages
684 across a diverse set of ecosystems (Figure S3) (Reid et al., 2005b). We find MEE values vary
685 between $2 - 6 \text{ m}^2 \text{ g}^{-1}$ for the FIREX-AQ smoke plumes and that the MEE increases
686 asymptotically as a function of smoke age (Figure 7). Our observations indicate MEE approaches
687 the mean from previous studies as the smoke rapidly evolves in the early hours after emission.
688 The rate at which MEE increases with smoke age is variable among the fires included in this
689 analysis and does not appear to depend on the plume PM concentration. Our range of MEEs for
690 smoke plumes from Western US wildland fires using high-resolution in situ measurements is
691 larger than what has been observed in previous studies. Our results emphasize the variability that
692 can occur in smoke MEE, and suggest that the top-down approach is likely more sensitive to
693 MEE than previous studies imply. The use of a constant MEE could lead to a high bias for fires
694 with lower excess PM concentrations and a low bias for fires with higher excess PM
695 concentrations, which would explain the trend we see in Figure 5 where FEER underestimates
696 fire E_{PM} from the most actively burning fires and overestimates E_{PM} from smaller, weaker fires.

697 **3.2.2.2 Instantaneous Observations of FRP and AOD**

698 FEER uses daytime MODIS FRP and AOD observations to derive C_e and assumes that
699 the FRP at the time of observation is directly related to the smoke plume AOD. However, fires
700 have a clear, ecosystem dependent diurnal cycle with the time of peak fire activity depending on
701 the specific landcover, geographic location, elevation, slope, aspect, and type of fire (e.g.,
702 wildland, prescribed, crown, surface). FRP observations represent the instantaneous fuel
703 consumption and corresponding emissions of a given fire, but AOD observations represent the

704 total mass of aerosols emitted by a fire, including the time period when the fire was active before
705 the satellite overpass time. The variability in FRP that occurs over the course of a day has a clear
706 impact on the total mass of smoke particles in the plume as a function of downwind distance
707 from the fire and wind speed, but polar orbiters, like MODIS do not have the temporal resolution
708 to quantify this relationship. As a result, C_e derived using FRP and AOD observed after the peak
709 in diurnal fire activity are likely overestimated, while C_e derived using FRP and AOD observed
710 prior to the peak may be slightly underestimated. The exact nature and magnitude of the potential
711 bias would depend on a specific fire's diurnal cycle and the age of the smoke captured in the
712 satellite observations of AOD. With respect to the calculation of E_{PM} using a predetermined C_e ,
713 the time offset between MODIS overpass times and peak diurnal fire activity could similarly
714 cause a bias. E_{PM} could potentially be biased high or low if the satellite overpass time occurred
715 either before or after the peak in diurnal fire activity, and if the observed FRP was higher or
716 lower than the daily average FRP.

717 A recent study by Mota and Wooster (2018) demonstrated fire emission rates can be
718 calculated at a high temporal and spatial resolution (hourly and $0.05^\circ \times 0.05^\circ$, respectively) using
719 a top-down approach that relies on geostationary satellite observations of FRP from the Spinning
720 Enhanced Visible and InfraRed Imager (SEVIRI) to avoid bias caused by inadequate sampling of
721 a fire's diurnal cycle. We compare geostationary satellite observations of FRP from GOES that
722 match the overpass times of MODIS with the average of all FRP observations over the course of
723 a day for each fire to investigate potential bias in E_{PM} estimates from FEER. The Western US
724 wildland fires sampled during FIREX-AQ exhibited peak fire activity from 3-6 pm local time
725 (Pacific daylight time, UTC-7) (Wiggins et al., 2020). Meanwhile, local MODIS overpass times
726 are $\sim 10:30$ am for the Terra satellite and $\sim 1:20$ pm for Aqua. We find average GOES FRP at the
727 time of the MODIS overpasses is double the daily average FRP from GOES (Figure S4), which
728 could be partially responsible for the overestimation in FEER E_{PM} estimates for smaller fires that
729 we see in Figure 5.

730 **4 Summary and Conclusions**

731 We present a comprehensive evaluation of total carbon and aerosol emission rate
732 estimates computed using the methodologies and assumptions that are commonly employed by
733 global inventories used by models. These emissions inventories have the monumental task of
734 capturing the composition, magnitude, and temporal variability of fire emissions from nearly
735 every ecosystem on Earth. They are critical for the representation of wildland fires in large scale
736 models, and only recently have sufficiently comprehensive observational datasets become
737 available to evaluate their performance. One such study is the joint NASA/NOAA FIREX-AQ
738 airborne mission that took place in 2019. Here, we extend the methods and assumptions
739 employed by emissions inventories to develop state-of-the-art, high-resolution emission rate
740 estimates for each of the western FIREX-AQ fires, which are based on detailed information
741 garnered from ground, airborne, and satellite assets.

742 We discover excellent agreement between the high-resolution emission rate estimates
743 calculated using integrated airborne in situ and lidar observations and the high-resolution top-
744 down (HSRL-GOES) and bottom-up (Fuel2Fire) estimates at unusually high sub-plume
745 spatiotemporal resolution. While there is considerable scatter in the one-to-one plots comparing
746 Fuel2Fire to the airborne in situ data, the emissions rate estimates for both total carbon and PM
747 are not consistently biased between these methodological approaches. HSRL-GOES appears to
748 slightly overestimate E_{PM} toward the lower end of the observed range of variability (which

749 appears to also scale with FRP). Emission rate estimates calculated using the lower resolution
750 global fire emissions inventories, FEER and GFED, have a weaker relationship with the high-
751 resolution approaches and show evidence of systematic bias, which is most apparent for GFED.

752 We discuss, in detail, the key assumptions employed by bottom-up approaches and
753 conclude that the strong performance of the Fuel2Fire inventory stems from detailed information
754 about fuel type and loading that are parameterized with significant uncertainty in the global
755 inventories. In addition, we note that the high-temporal resolution of the Fuel2Fire dataset also
756 allows it to capture the entire diurnal cycle of the fire activity, which also serves to improve its
757 predictive skill. This hints that the high temporal resolution of geostationary satellite
758 observations of FRP could be used to correct the bias caused by satellite overpass times. With
759 respect to top-down approaches, we find a larger range in MEE for this small subset of Western
760 US fires than what has been reported in a compilation of previous studies that includes MEE
761 from fires in a diverse selection of global ecosystems. The high resolution top-down approach
762 (HSRL-GOES) allowed for the application of variable MEE obtained from sub-plume in situ
763 measurements. HSRL extinction measurements of the smoke plumes sampled during FIREX-AQ
764 combined with geostationary satellite observations of FRP offered an exceptionally detailed
765 measure of AOD and FRP associated with the smoke plume. The use of a constant MEE to
766 convert AOD to PM in top-down approaches combined with bias from assumptions related to
767 instantaneous observations of FRP and AOD are likely responsible for the underestimation in
768 FEER E_{PM} for larger fires and overestimation for smaller fires.

769 Finally, it's important to note that it is not yet computationally practical or feasible for
770 global fire emissions inventories to achieve the level of complexity and detail in the high-
771 resolution approaches presented here. We use these approaches to investigate discrepancies
772 between top-down and bottom-up E_{PM} estimates for Western US wildland fires. However, this
773 collection of fires represents only a small subset of the total number of fires that burn every year
774 in the Western US and may not be a perfect representation of the complexity that can exist in fire
775 emissions. In short, we have the luxury of evaluating the skill of the global emissions inventories
776 for a small subset of wildland fires for which we have unprecedentedly comprehensive data, but
777 we would be wise to remember that the goal of global emissions inventories is to represent all
778 fires reasonably well rather than to represent a few fires perfectly. Consequently, it may be
779 premature to adopt new values for, e.g., the smoke emission coefficient based solely on the
780 FIREX-AQ dataset. Our analysis does emphasize areas of large uncertainty, however, that may
781 be improved. One is the estimate of fuel type and loading that likely contributes to the scatter we
782 see in the bottom-up emission rate estimates from GFED and Fuel2Fire. Burned area and aerosol
783 mass emission factors do not appear to be large sources of uncertainty as there is good agreement
784 seen for both GFED and Fuel2Fire for both of these metrics. The importance of the high-
785 temporal resolution observations of both FRP and smoke AOD afforded by the geostationary
786 satellites currently in orbit cannot be overstated, as a lack of complete orbital coverage is also
787 likely to be a strong contributor to the inventory emissions underestimates. The use of a constant
788 MEE to convert AOD to PM should be revisited in light of the much higher variability we find in
789 MEE observations for such a limited number of fires, which accentuates the need for additional
790 measurements of this key variable. In summary, both top-down and bottom-up global fire
791 emissions inventories suffer from assumptions that may hold true in the aggregate, but break
792 down on an individual fire basis. The strong agreement that we show here between the high-
793 resolution approaches holds promise for future fire emissions inventories as advances in remote

794 sensing, improved computational efficiency, and a more complete understanding of fire behavior
795 begin to offer opportunities to increase the accuracy and resolution of global fire inventories.

796 **Appendix: List of Variables and Common Units**

797 *Note: units are included here only as examples and do not consider any unit conversions that*
798 *may be necessary for the equations given in the text.*

799	α_t	HSRL extinction coefficient	km^{-1}
800	$A_{P,FEER}$	FEER pixel area	km^2
801	$A_{P,GFED}$	GFED pixel area	km^2
802	AOD	Aerosol optical depth	unitless
803	β	HSRL backscatter ratio	$\text{km}^{-1} \text{sr}^{-1}$
804	BA	Burned area	m^2
805	CC	Combustion completeness	%
806	C_e	Smoke emission coefficient	gPM MW^{-1}
807	ΔC	Excess mass concentration of C	$\mu\text{gC m}^{-3}$
808	ΔPM	Excess mass concentration of PM	$\mu\text{gPM m}^{-3}$
809	Δt	HSRL curtain pixel width	s
810	Δz	HSRL curtain pixel height	m
811	E_C	Emission rate of total carbon	kgC s^{-1}
812	\widehat{E}_C	Area-normalized emission rate of total carbon	$\text{kgC m}^{-2} \text{s}^{-1}$
813	E_{PM}	Emission rate of total PM	kgPM s^{-1}
814	\widehat{E}_{PM}	Area-normalized emission rate of total PM	$\text{kgPM m}^{-2} \text{s}^{-1}$
815	EF_{PM}	Particle mass emissions factor	$\text{gPM kg biomass consumed}^{-1}$
816	F_C	Mass fraction of carbon in the fuel	$\text{gC kg biomass consumed}^{-1}$
817	FL	Fuel loading	g biomass m^{-2}
818	FRP	Fire radiative power	MW
819	H	Plume vertical thickness	m
820	\overline{MEE}	Particle mass extinction efficiency	$\text{m}^2 \text{g}^{-1}$
821	\overline{MEE}	Aircraft transect-average MEE	$\text{m}^2 \text{g}^{-1}$
822	PM	Particle mass concentration	$\mu\text{gPM m}^{-3}$
823	\overline{GS}	Transect-average DC-8 aircraft ground speed	m s^{-1}
824	\overline{WS}	Aircraft transect-average wind speed	m s^{-1}

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835 **Conflict of Interest**

836 The authors declare no conflicts of interest or competing interests.

837 **Author Contributions**

838 EBW formal analysis and investigation. EBW and RHM conceptualization and writing – original
839 draft. EBW, PCJ, GC, JPD, GSD, MF, EG, HG, HWH, HSH, CI, JLJ., JMK, JBN, AEP, CER,
840 KJS, MS, JPS, TJS, MAS, AS, ELW, and RHM data curation. JC and CW project
841 administration. All co-authors writing – reviewing and editing.

842 **References**

- 843 Abatzoglou, J. T., & Williams, A. P. (2016). Impact of anthropogenic climate change on wildfire
844 across western US forests. *Proceedings of the National Academy of Sciences of the United*
845 *States of America*, 113(42), 11770–11775. <https://doi.org/10.1073/pnas.1607171113>
- 846 Adachi, K., J. E. Dibb, E. Scheuer, J. M. Katich, J. P. Schwarz, A. E. Perring, B. Mediavilla, H.
847 Guo, P. Campuzano-Jost, J. L. Jimenez, J. Crawford, N. Oshima, M. Kajino, T. Kinase, L.
848 Kleinman, A. J. Sedlacek III, R. J. Yokelson, & P. R. Buseck (2021), Fine ash particles as a
849 major aerosol component in biomass burning smoke, submitted to *Journal of Geophysical*
850 *Research: Atmospheres*.
- 851 Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J.
852 D. & Wennberg, P. O. (2011). Emission factors for open and domestic biomass burning for
853 use in atmospheric models. *Atmospheric Chemistry and Physics*, 11(9), 4039–4072.
854 <https://doi.org/10.5194/acp-11-4039-2011>
- 855 Akagi, S. K., Craven, J. S., Taylor, J. W., McMeeking, G. R., Yokelson, R. J., Burling, I. R.,
856 Urbanski, S. P., Wold, C. E., Seinfeld, J. H., Coe, H. & Alvarado, M.J. (2012). Evolution of
857 trace gases and particles emitted by a chaparral fire in California. *Atmospheric Chemistry*
858 *and Physics*, 12(3), 1397–1421. <https://doi.org/10.5194/acp-12-1397-2012>
- 859 Anderson, T. L. & Ogren, J. A. (1998). Determining aerosol radiative properties using the TSI
860 3563 integrating nephelometer. *Aerosol Science and Technology*, 29(1), 57-69.
861 <https://doi.org/10.1080/02786829808965551>
- 862 Andreae, M. O. (2019). Emission of trace gases and aerosols from biomass burning – an updated
863 assessment, *Atmospheric Chemistry and Physics*, 19(13), 8523–8546.
864 <https://doi.org/10.5194/acp-19-8523-2019>, 2019
- 865 Andreae, M. O., & Merlet, P. (2001). Emission of trace gases and aerosols from biomass
866 burning. *Global Biogeochemical Cycles*, 15(4), 955–966.
867 <https://doi.org/10.1029/2000GB001382>
- 868 Boschetti, L., Roy, D. P., Giglio, L., Huang, H., Zubkova, M., & Humber, M. L. (2019). Global
869 validation of the collection 6 MODIS burned area product. *Remote Sensing of Environment*,
870 235, 111490. <https://doi.org/10.1016/j.rse.2019.111490>
- 871 Burling, I. R., Yokelson, R. J., Akagi, S. K., Urbanski, S. P., Wold, C. E., Griffith, D. W.,
872 Johnson, T. J., Reardon, J. & Weise, D. R. (2011). Airborne and ground-based

- 873 measurements of the trace gases and particles emitted by prescribed fires in the United
874 States. *Atmospheric Chemistry and Physics*, 11(23), 12197-12216.
875 <https://doi.org/10.5194/acp-11-12197-2011>
- 876 Burling, I. R., Yokelson, R. J., Griffith, D. W., Johnson, T. J., Veres, P., Roberts, J. M.,
877 Warneke, C., Urbanski, S. P., Reardon, J., Weise, D. R. & Hao, W. M. (2010). Laboratory
878 measurements of trace gas emissions from biomass burning of fuel types from the
879 southeastern and southwestern United States. *Atmospheric Chemistry and Physics*, 10(22),
880 11115-11130. <https://doi.org/10.5194/acp-10-11115-2010>
- 881 Canagaratna, M. R., Jayne, J. T., Jimenez, J. L., Allan, J. D., Alfarra, M. R., Zhang, Q., Onasch,
882 T. B., Drewnick, F., Coe, H., Middlebrook, A. & Delia, A. (2007). Chemical and
883 microphysical characterization of ambient aerosols with the aerodyne aerosol mass
884 spectrometer. *Mass Spectrometry Reviews*, 26(2), 85-222.
885 <https://doi.org/10.1002/mas.20115>
- 886 Cappa, C. D., Lim, C. Y., Hagan, D. H., Coggon, M., Koss, A., Sekimoto, K., Gouw, J. D.,
887 Onasch, T. B., Warneke, C. & Kroll, J. H. (2020). Biomass-burning-derived particles from a
888 wide variety of fuels-Part 2: Effects of photochemical aging on particle optical and
889 chemical properties. *Atmospheric Chemistry and Physics*, 20(14), 8511–8532.
890 <https://doi.org/10.5194/acp-20-8511-2020>
- 891 Carter, T. S., Heald, C. L., Jimenez, J. L., Campuzano-Jost, P., Kondo, Y., Moteki, N., Schwarz,
892 J. P., Wiedinmyer, C., Darmenov, A. S., Silva, A. M. D. & Kaiser, J. W. (2020). How
893 emissions uncertainty influences the distribution and radiative impacts of smoke from fires
894 in North America. *Atmospheric Chemistry and Physics*, 20(4), 2073–2097.
895 <https://doi.org/10.5194/acp-20-2073-2020>
- 896 Darmenov, A. S., & da Silva, A. (2013). *The Quick Fire Emissions Dataset (QFED) -*
897 *Documentation of versions 2.1, 2.2, and 2.4, NASA Technical Report Series on Global*
898 *Modeling and Data Assimilation, Volume 32.*
- 899 DeCarlo, P. F., Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C., Gonin,
900 M., Fuhrer, K., Horvath, T., Docherty, K. S. & Worsnop, D. R. (2006). Field-deployable,
901 high-resolution, time-of-flight aerosol mass spectrometer. *Analytical Chemistry*, 78(24),
902 8281-8289. <https://doi.org/10.1021/ac061249n>
- 903 Dennison, P. E., Brewer, S. C., Arnold, J. D., & Moritz, M. A. (2014). Large wildfire trends in
904 the western United States, 1984-2011. *Geophysical Research Letters*, 41(8), 2928–2933.
905 <https://doi.org/10.1002/2014GL059576>
- 906 Freeborn, P. H., Wooster, M. J., Hao, W. M., Ryan, C. A., Nordgren, B. L., Baker, S. P., &
907 Ichoku, C. (2008). Relationships between energy release, fuel mass loss, and trace gas an
908 aerosol emissions during laboratory biomass fires. *Journal of Geophysical Research:*
909 *Atmospheres*, 113, D01301. <https://doi.org/10.1029/2007JD008679>
- 910 Freeborn, P. H., Wooster, M. J., Roy, D. P. & Cochrane, M. A. (2014). Quantification of MODIS
911 fire radiative power (FRP) measurement uncertainty for use in satellite-based active fire

- 912 characterization and biomass burning estimation. *Geophysical Research Letters*, 41(6),
913 1988-1994. <https://doi.org/10.1002/2013GL059086>
- 914 French, N. H. F., Goovaerts, P., & Kasischke, E. S. (2004). Uncertainty in estimating carbon
915 emissions from boreal forest fires. *Journal of Geophysical Research: Atmospheres*, 109(14),
916 14–22. <https://doi.org/10.1029/2003JD003635>
- 917 Fusco, E. J., Abatzoglou, J. T., Balch, J. K., Finn, J. T., & Bradley, B. A. (2016). Quantifying the
918 human influence on fire ignition across the western USA. *Ecological Applications*, 26(8),
919 2388–2399. <https://doi.org/10.1002/eap.1395>
- 920 Garofalo, L. A., Pothier, M. A., Levin, E. J. T., Campos, T., Kreidenweis, S. M., & Farmer, D.
921 K. (2019). Emission and Evolution of Submicron Organic Aerosol in Smoke from Wildfires
922 in the Western United States. *ACS Earth and Space Chemistry*, 3(7), 1237–1247.
923 <https://doi.org/10.1021/acsearthspacechem.9b00125>
- 924 Giglio, L., Csiszar, I., & Justice, C. O. (2006). Global distribution and seasonality of active fires
925 as observed with the Terra and Aqua Moderate Resolution Imaging Spectroradiometer
926 (MODIS) sensors. *Journal of Geophysical Research: Biogeosciences*, 111(2), 2016.
927 <https://doi.org/10.1029/2005JG000142>
- 928 Giglio, L., Boschetti, L., Roy, D. P., Humber, M. L. & Justice, C. O. (2018). The Collection 6
929 MODIS burned area mapping algorithm and product. *Remote Sensing of Environment*, 217,
930 72-85. <https://doi.org/10.1016/j.rse.2018.08.005>
- 931 Goldammer, J. G., Statheropoulos, M., & Andreae, M. O. (2008). Impacts of Vegetation Fire
932 Emissions on the Environment, Human Health, and Security: A Global Perspective.
933 *Developments in Environmental Science*, 8, 3-36. [https://doi.org/10.1016/S1474-](https://doi.org/10.1016/S1474-8177(08)00001-6)
934 [8177\(08\)00001-6](https://doi.org/10.1016/S1474-8177(08)00001-6)
- 935 Guo, X., Shang, Y., Lv, Y., Bai, H. & Ma, Q. (2021). Suspect Screening of Fentanyl Analogs
936 Using Matrix-Assisted Ionization and a Miniature Mass Spectrometer with a Custom
937 Expandable Mass Spectral Library. *Analytical Chemistry*, 93(29), 10152-10159.
938 <https://doi.org/10.1021/acs.analchem.1c01117>
- 939 Harvey, B. J. (2016). Human-caused climate change is now a key driver of forest fire activity in
940 the western United States. *Proceedings of the National Academy of Sciences of the United*
941 *States of America*, 113(42), 11649-11650. <https://doi.org/10.1073/pnas.1612926113>
- 942 Hodshire, A. L., Bian, Q., Ramnarine, E., Lonsdale, C. R., Alvarado, M. J., Kreidenweis, S. M.,
943 Jathar, S. H. & Pierce, J. R. (2019). More Than Emissions and Chemistry: Fire Size,
944 Dilution, and Background Aerosol Also Greatly Influence Near-Field Biomass Burning
945 Aerosol Aging. *Journal of Geophysical Research: Atmospheres*, 124(10), 5589–5611.
946 <https://doi.org/10.1029/2018JD029674>
- 947 Ichoku, C., & Ellison, L. (2014). Global top-down smoke-aerosol emissions estimation using
948 satellite fire radiative power measurements. *Atmospheric Chemistry and Physics*, 14(13),
949 6643–6667. <https://doi.org/10.5194/acp-14-6643-2014>

- 950 Ichoku, Charles, & Kaufman, Y. J. (2005). A method to derive smoke emission rates from
 951 MODIS fire radiative energy measurements. *IEEE Transactions on Geoscience and Remote*
 952 *Sensing*, 43, 2636–2649. <https://doi.org/10.1109/TGRS.2005.857328>
- 953 Ichoku, Charles, Giglio, L., Wooster, M. J., & Remer, L. A. (2008). Global characterization of
 954 biomass-burning patterns using satellite measurements of fire radiative energy. *Remote*
 955 *Sensing of Environment*, 112(6), 2950–2962. <https://doi.org/10.1016/j.rse.2008.02.009>
- 956 Kaiser, J. W., Flemming, J., Schultz, M. G., Suttie, M., & Wooster, M. J. (2009). *The MACC*
 957 *Global Fire Assimilation System: First Emission Products (GFASv0)*. Retrieved from
 958 <http://www.ecmwf.int/publications/>
- 959 Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J. J.,
 960 Razingier, M., Schultz, M. G., Suttie, M. & Van Der Werf, G. R. (2012). Biomass burning
 961 emissions estimated with a global fire assimilation system based on observed fire radiative
 962 power. *Biogeosciences*, 9(1), 527–554. <https://doi.org/10.5194/bg-9-527-2012>
- 963 Keane, R. E., Herynk, J. M., Toney, C., Urbanski, S. P., Lutes, D. C. & Ottmar, R. D. (2013).
 964 Evaluating the performance and mapping of three fuel classification systems using Forest
 965 Inventory and Analysis surface fuel measurements. *Forest Ecology and Management*, 305,
 966 248-263. <https://doi.org/10.1016/j.foreco.2013.06.001>
- 967 Kennedy, M. C., Prichard, S. J., McKenzie, D., & French, N. H. F. (2020). Quantifying how
 968 sources of uncertainty in combustible biomass propagate to prediction of wildland fire
 969 emissions. *International Journal of Wildland Fire*, 29(9), 793–806.
 970 <https://doi.org/10.1071/WF19160>
- 971 Larkin, N. K., O'Neill, S. M., Solomon, R., Raffuse, S., Strand, T., Sullivan, D. C., Krull, C.,
 972 Rorig, M., Peterson, J. & Ferguson, S. A. (2009). The BlueSky smoke modeling framework.
 973 *International Journal of Wildland Fire*, 18(8), 906–920. <https://doi.org/10.1071/WF07086>
- 974 Larkin, N. K., Raffuse, S. M., & Strand, T. M. (2014). Wildland fire emissions, carbon, and
 975 climate: U.S. emissions inventories. *Forest Ecology and Management*, 317, 61–69.
 976 <https://doi.org/10.1016/j.foreco.2013.09.012>
- 977 Larsen, A. E., Reich, B. J., Ruminski, M., & Rappold, A. G. (2018). Impacts of fire smoke
 978 plumes on regional air quality, 2006-2013. *Journal of Exposure Science and Environmental*
 979 *Epidemiology*, 28(4), 319-327. <https://doi.org/10.1038/s41370-017-0013-x>
- 980 Li, F., Val Martin, M., Andreae, M. O., Arneth, A., Hantson, S., Kaiser, J. W., Lasslop, G., Yue,
 981 C., Bachelet, D., Forrest, M. & Kluzek, E. (2019). Historical (1700–2012) global multi-
 982 model estimates of the fire emissions from the Fire Modeling Intercomparison Project
 983 (FireMIP). *Atmospheric Chemistry and Physics*, 19(19), 12545–12567.
 984 <https://doi.org/10.5194/acp-19-12545-2019>
- 985 Liu, T., Mickley, L. J., Marlier, M. E., DeFries, R. S., Khan, M. F., Latif, M. T., & Karambelas,
 986 A. (2020). Diagnosing spatial biases and uncertainties in global fire emissions inventories:
 987 Indonesia as regional case study. *Remote Sensing of Environment*, 237, 111557.

- 988 <https://doi.org/10.1016/j.rse.2019.111557>
- 989 Liu, Y. (2004). Variability of wildland fire emissions across the contiguous United States.
990 *Atmospheric Environment*, 38(21), 3489–3499.
991 <https://doi.org/10.1016/j.atmosenv.2004.02.004>
- 992 May, A. A., McMeeking, G. R., Lee, T., Taylor, J. W., Craven, J. S., Burling, I., Sullivan, A. P.,
993 Akagi, S., Collett Jr., J. L., Flynn, M. & Coe, H. (2014). Aerosol emissions from prescribed
994 fires in the United States: A synthesis of laboratory and aircraft measurements. *Journal of*
995 *Geophysical Research: Atmospheres*, 119(20), 11826–11849.
996 <https://doi.org/10.1002/2014JD021848>
- 997 McClure, C. D., Lim, C. Y., Hagan, D. H., Kroll, J. H., & Cappa, C. D. (2020). Biomass-
998 burning-derived particles from a wide variety of fuels - Part 1: Properties of primary
999 particles. *Atmospheric Chemistry and Physics*, 20(3), 1531–1547.
1000 <https://doi.org/10.5194/acp-20-1531-2020>
- 1001 McKenzie, D., O'Neill, S. M., Larkin, N. K., & Norheim, R. A. (2006). Integrating models to
1002 predict regional haze from wildland fire. *Ecological Modelling*, 199(3), 278–288.
1003 <https://doi.org/10.1016/j.ecolmodel.2006.05.029>
- 1004 McKenzie, D., French, N. H. F., & Ottmar, R. D. (2012). National database for calculating fuel
1005 available to wildfires. *Eos, Transactions American Geophysical Union*, 93(6), 57–58.
1006 <https://doi.org/10.1029/2012EO060002>
- 1007 McMeeking, G. R., Kreidenweis, S. M., Baker, S., Carrico, C. M., Chow, J. C., Collett Jr., J. L.,
1008 Hao, W. M., Holden, A. S., Kirchstetter, T. W., Malm, W. C. & Moosmüller, H. (2009).
1009 Emissions of trace gases and aerosols during the open combustion of biomass in the
1010 laboratory. *Journal of Geophysical Research: Atmospheres*, 114, D19210.
1011 <https://doi.org/10.1029/2009JD011836>
- 1012 Mell, W. E., Manzello, S. L., Maranghides, A., Butry, D., & Rehm, R. G. (2010). The wildland -
1013 urban interface fire problem - current approaches and research needs. *International Journal*
1014 *of Wildland Fire*, 19(2), 238-251. <https://doi.org/10.1071/WF07131>
- 1015 Moore, R. H., Wiggins, E. B., Ahern, A. T., Zimmerman, S., Montgomery, L., Campuzano Jost,
1016 P., Robinson, C. E., Ziemba, L. D., Winstead, E. L., Anderson, B. E. & Brock, C.A. (2021).
1017 Sizing response of the Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) and Laser
1018 Aerosol Spectrometer (LAS) to changes in submicron aerosol composition and refractive
1019 index. *Atmospheric Measurement Techniques*, 14(6), 4517-4542.
1020 <https://doi.org/10.5194/amt-14-4517-2021>
- 1021 Mota, B., & Wooster, M. J. (2018). A new top-down approach for directly estimating biomass
1022 burning emissions and fuel consumption rates and totals from geostationary satellite fire
1023 radiative power (FRP). *Remote Sensing of Environment*, 206, 45–62.
1024 <https://doi.org/10.1016/j.rse.2017.12.016>
- 1025 Nikonovas, T., North, P. R. J., & Doerr, S. H. (2017). Particulate emissions from large North

- 1026 American wildfires estimated using a new top-down method. *Atmospheric Chemistry and*
 1027 *Physics*, 17(10), 6423–6438. <https://doi.org/10.5194/acp-17-6423-2017>
- 1028 Noss, R. F., Franklin, J. F., Baker, W. L., Schoennagel, T., & Moyle, P. B. (2006). Managing
 1029 fire-prone forests in the western United States. *Frontiers in Ecology and the Environment*,
 1030 4(9), 481–487. [https://doi.org/https://doi.org/10.1890/1540-](https://doi.org/https://doi.org/10.1890/1540-9295(2006)4[481:MFFITW]2.0.CO;2)
 1031 [9295\(2006\)4\[481:MFFITW\]2.0.CO;2](https://doi.org/https://doi.org/10.1890/1540-9295(2006)4[481:MFFITW]2.0.CO;2)
- 1032 Ottmar, R. D., Sandberg, D. V., Riccardi, C. L., & Prichard, S. J. (2007). An overview of the
 1033 Fuel Characteristic Classification System — Quantifying, classifying, and creating fuelbeds
 1034 for resource planning This article is one of a selection of papers published in the Special
 1035 Forum on the Fuel Characteristic Classification System. *Canadian Journal of Forest*
 1036 *Research*, 37(12), 2383–2393. <https://doi.org/10.1139/X07-077>
- 1037 Pan, X., Ichoku, C., Chin, M., Bian, H., Darmenov, A., Colarco, P., Ellison, L., Kucsera, T.,
 1038 Silva, A. D., Wang, J. & Oda, T. (2020). Six global biomass burning emission datasets:
 1039 intercomparison and application in one global aerosol model. *Atmospheric Chemistry and*
 1040 *Physics*, 20(2), 969–994. <https://doi.org/10.5194/acp-20-969-2020>
- 1041 Pettinari, M. L., & Chuvieco, E. (2016). Generation of a global fuel data set using the Fuel
 1042 Characteristic Classification System. *Biogeosciences*, 13(7), 2061–2076.
 1043 <https://doi.org/10.5194/bg-13-2061-2016>
- 1044 Prichard, S. J., Kennedy, M. C., Andreu, A. G., Eagle, P. C., French, N. H., & Billmire,
 1045 M. (2019). Next-generation biomass mapping for regional emissions and carbon
 1046 inventories: Incorporating uncertainty in wildland fuel characterization. *Journal of*
 1047 *Geophysical Research: Biogeosciences*, 124(12), 3699–3716.
 1048 <https://doi.org/10.1071/WF19066>.
- 1049 Prichard, S. J., O’neill B, S. M., Eagle, P., Andreu, A. G., Drye, B., Dubowy, J., Urbanski, S., &
 1050 Strand, T. (2020). Wildland fire emission factors in North America: synthesis of existing
 1051 data, measurement needs and management applications. *International Journal of Wildland*
 1052 *Fire*, 29(2), 132–147. <https://doi.org/10.1071/WF19066>
- 1053 Randerson, J. T., Chen, Y., Van Der Werf, G. R., Rogers, B. M., & Morton, D. C. (2012). Global
 1054 burned area and biomass burning emissions from small fires. *Journal of Geophysical*
 1055 *Research: Biogeosciences*, 117, G04012. <https://doi.org/10.1029/2012JG002128>
- 1056 Reid, J. S., Koppmann, R., Eck, T. F., & Eleuterio, D. P. (2005a). A review of biomass burning
 1057 emissions part II: Intensive physical properties of biomass burning particles. *Atmospheric*
 1058 *Chemistry and Physics*, 5(3), 799–825. <https://doi.org/10.5194/acp-5-799-2005>
- 1059 Reid, J.S., Eck, T.F., Christopher, S.A., Koppmann, R., Dubovik, O., Eleuterio, D.P., Holben,
 1060 B.N., Reid, E.A. & Zhang, J. (2005b). A review of biomass burning emissions part III:
 1061 Intensive optical properties of biomass burning particles. *Atmospheric Chemistry and*
 1062 *Physics*, 5(3), 827–849, <https://doi.org/10.5194/acp-5-827-2005>
- 1063 Sachse, G. W., Collins, J. E. Jr., Hill, G. F., Wade, L. O., Burney, L. G., & Ritter, J. A. (1991).
 1064 Airborne tunable diode laser sensor for high-precision concentration and flux measurements

- 1065 of carbon monoxide and methane. *Measurement of Atmospheric Gases, 1433*, 157–166.
1066 <https://doi.org/10.1117/12.46162>
- 1067 Sandberg, D. V., Ottmar, R. D., & Cushon, G. H. (2001). Characterizing fuels in the 21st
1068 Century. *International Journal of Wildland Fire, 10*(4), 381–387.
1069 <https://doi.org/10.1071/wf01036>
- 1070 Santín, C., Doerr, S. H., Preston, C. M., & González-Rodríguez, G. (2015). Pyrogenic organic
1071 matter production from wildfires: a missing sink in the global carbon cycle. *Global Change*
1072 *Biology, 21*(4), 1621–1633. <https://doi.org/10.1111/gcb.12800>
- 1073 Schmidt, C. (2019). Monitoring Fires with the GOES-R Series. In *The GOES-R Series: A New*
1074 *Generation of Geostationary Environmental Satellites* (pp. 145–163). Elsevier.
1075 <https://doi.org/10.1016/B978-0-12-814327-8.00013-5>
- 1076 Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke, K., Goldammer, J. G., Held,
1077 A. C., Pereira, J. M. & van Het Bolscher, M. (2008). Global wildland fire emissions from
1078 1960 to 2000. *Global Biogeochemical Cycles, 22*, GB2002.
1079 <https://doi.org/10.1029/2007GB003031>
- 1080 Schweizer, D., Preisler, H. K., & Cisneros, R. (2018). Assessing relative differences in smoke
1081 exposure from prescribed, managed, and full suppression wildland fire. *Air Quality,*
1082 *Atmosphere & Health, 12*(1), 87-95. <https://doi.org/10.1007/s11869-018-0633-x>
- 1083 Seiler, W., & Crutzen, P. J. (1980). Estimates of gross and net fluxes of carbon between the
1084 biosphere and the atmosphere from biomass burning. *Climatic Change, 2*(3), 207–247.
1085 <https://doi.org/10.1007/BF00137988>
- 1086 Seinfeld, J. H., & Pandis, S. N. (2006). Interaction of Aerosols with Radiation. In *Atmospheric*
1087 *Chemistry and Physics: from Air Pollution to Climate Change* (pp. 691–719). Hoboken,
1088 New Jersey: John Wiley & Sons.
- 1089 Shingler, T., Crosbie, E., Ortega, A., Shiraiwa, M., Zuend, A., Beyersdorf, A., Ziemba, L.,
1090 Anderson, B., Thornhill, L., Perring, A. E., Schwarz, J. P., Campazano-Jost, P., Day, D. A.,
1091 Jimenez, J. L., Hair, J. W., Mikoviny, T., Wisthaler, A., & Sorooshian, A. (2016). Airborne
1092 Characterization of Sub-saturated Aerosol Hygroscopicity and Dry Refractive Index from
1093 the Surface to 6.5 km during the SEAC⁴RS Campaign, *Journal Geophysical Research:*
1094 *Atmospheres, 121*(8), 4188–4210. <https://doi.org/10.1002/2015JD024498>
- 1095 Stavros, E. N., Abatzoglou, J., Larkin, N. K., McKenzie, D., & Steel, E. A. (2014). Climate and
1096 very large wildland fires in the contiguous western USA. *International Journal of Wildland*
1097 *Fire, 23*(7), 899-914. <https://doi.org/10.1071/WF13169>
- 1098 Stephens, S. L., & Ruth, L. W. (2005). Federal forest-fire policy in the United States. *Ecological*
1099 *Applications, 15*(2), 532-542. <https://doi.org/10.1890/04-0545>
- 1100 Surawski, N. C., Sullivan, A. L., Roxburgh, S. H., Meyer, C. P. M., & Polglase, P. J. (2016).
1101 Incorrect interpretation of carbon mass balance biases global vegetation fire emission

- 1102 estimates. *Nature Communications*, 7(1), 1–5. <https://doi.org/10.1038/ncomms11536>
- 1103 Susott, R. A., Ward, D. E., Babbitt, R. E., & Latham, D. J. (1991). The measurement of trace
1104 emissions and combustion characteristics for a mass fire In Levine, J. S. (Eds.), *Global*
1105 *Biomass Burning: Atmospheric, Climatic, and Biosphere Implications* (pp. 245-257).
1106 Cambridge, Massachusetts: The MIT Press.
- 1107 Susott, R. A., Olbu, G. J., Baker, S. P. & Ward, D. E. (1996). Carbon, Hydrogen, Nitrogen, and
1108 Thermogravimetric Analysis. In Levine, J. S. (Eds.), *Biomass Burning and Global Change:*
1109 *Remote sensing, Modeling and Inventory Development, and Biomass Burning in Africa*
1110 (Vol. 1, pp 249-259). Cambridge, Massachusetts: The MIT Press.
- 1111 Theobald, D. M., & Romme, W. H. (2007). Expansion of the US wildland-urban interface.
1112 *Landscape and Urban Planning*, 83(4), 340–354.
1113 <https://doi.org/10.1016/j.landurbplan.2007.06.002>
- 1114 Urbanski, S. P., Hao, W. M., & Nordgren, B. (2011). The wildland fire emission inventory:
1115 Western United States emission estimates and an evaluation of uncertainty. *Atmospheric*
1116 *Chemistry and Physics*, 11(24), 12973–13000. <https://doi.org/10.5194/acp-11-12973-2011>
- 1117 Ward, D. E., & Radke, L. F. (1993). Emissions measurements from vegetation fires: A
1118 comparative evaluation of methods and results. In P. J. Crutzen & J. G. Goldammer (Eds.),
1119 *Fire in the Environment: The Ecological, Atmospheric, and Climatic Importance of*
1120 *Vegetation Fires* (pp. 53–76). Chichester, England: John Wiley & Sons.
- 1121 Weaver, H. (1974). Effects of fire on temperate forests: western United States. In C. E.
1122 Kozlowski & T. T. Ahlgren (Eds.), *Fire and Ecosystems* (pp. 279–317). New York, New
1123 York: Academic Press.
- 1124 Van Der Werf, G. R., Randerson, J. T., Giglio, L., Van Leeuwen, T. T., Chen, Y., Rogers, B. M.,
1125 Mu, M., Van Marle, M. J., Morton, D. C., Collatz, G. J. & Yokelson, R.J. (2017). Global
1126 fire emissions estimates during 1997–2016. *Earth System Science Data*, 9(2), 697–720.
1127 <https://doi.org/10.5194/essd-9-697-2017>
- 1128 van Leeuwen, T. T., van der Werf, G. R., Hoffmann, A. A., Detmers, R. G., Rücker, G., French,
1129 N. H. F., Archibald, S., Carvalho Jr., J. A., Cook, G. D., de Groot, W. J., Hély, C.,
1130 Kasischke, E. S., Kloster, S., McCarty, J. L., Pettinari, M. L., Savadogo, P., Alvarado, E. C.,
1131 Boschetti, L., Manuri, S., Meyer, C. P., Siegert, F., Trollope, L. A., & Trollope, W. S. W.
1132 (2014). Biomass burning fuel consumption rates: a field measurement database,
1133 *Biogeosciences*, 11(24), 7305–7329, <https://doi.org/10.5194/bg-11-7305-2014>
- 1134 Vay, S. A., Woo, J. H., Anderson, B. E., Thornhill, K. L., Blake, D. R., Westberg, D. J., Kiley,
1135 C. M., Avery, M. A., Sachse, G. W., Streets, D. G., & Tsutsumi, Y. (2003). Influence of
1136 regional-scale anthropogenic emissions on CO₂ distributions over the western North
1137 Pacific. *Journal of Geophysical Research: Atmospheres*, 108, 8801.
1138 <https://doi.org/10.1029/2002JD003094>
- 1139 Virkkula, A. (2010). Correction of the calibration of the 3-wavelength Particle Soot Absorption

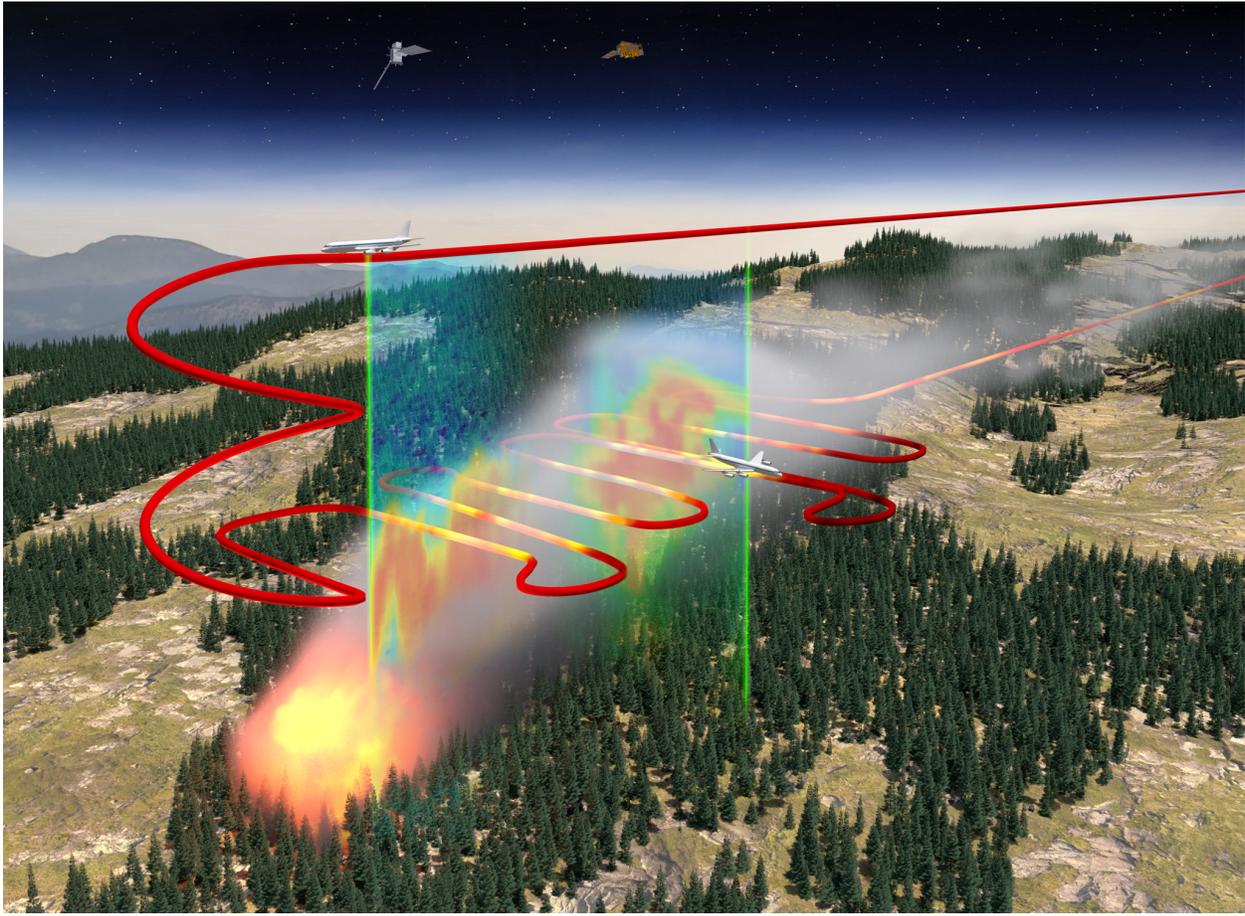
- 1140 Photometer (3λ PSAP). *Aerosol Science and Technology*, 44(8), 706-712.
1141 <https://doi.org/10.1080/02786826.2010.482110>
- 1142 Westerling, A. L., Gershunov, A., Brown, T. J., Cayan, D. R., & Dettinger, M. D. (2003).
1143 Climate and Wildfire in the Western United States. *Bulletin of the American Meteorological*
1144 *Society*, 84(5), 595–604. <https://doi.org/10.1175/BAMS-84-5-595>
- 1145 Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., &
1146 Soja, A. J. (2011). The Fire INventory from NCAR (FINN): A high resolution global model
1147 to estimate the emissions from open burning. *Geoscientific Model Development*, 4(3), 625–
1148 641. <https://doi.org/10.5194/gmd-4-625-2011>
- 1149 Wiedinmyer, C., Quayle, B., Geron, C., Belote, A., McKenzie, D., Zhang, X., O’Neill, S. &
1150 Wynne, K. K. (2006). Estimating emissions from fires in North America for air quality
1151 modeling. *Atmospheric Environment*, 40(19), 3419–3432.
1152 <https://doi.org/10.1016/j.atmosenv.2006.02.010>
- 1153 Wiggins, E. B., Soja, A. J., Gargulinski, E., Halliday, H. S., Pierce, R. B., Schmidt, C. C.,
1154 Nowak, J. B., DiGangi, J. P., Diskin, G. S., Katich, J. M. & Perring, A. E. (2020). High
1155 Temporal Resolution Satellite Observations of Fire Radiative Power Reveal Link Between
1156 Fire Behavior and Aerosol and Gas Emissions. *Geophysical Research Letters*, 47(23),
1157 e2020GL090707. <https://doi.org/10.1029/2020GL090707>
- 1158 Wiggins, E. B., Andrews, A., Sweeney, C., Miller, J. B., Miller, C. E., Veraverbeke, S.,
1159 Commane, R., Wofsy, S., Henderson, J. M. & Randerson, J. T. (2021). Boreal forest fire
1160 CO and CH₄ emission factors derived from tower observations in Alaska during the extreme
1161 fire season of 2015. *Atmospheric Chemistry and Physics*, 21(11), 8557–8574.
1162 <https://doi.org/10.5194/acp-21-8557-2021>
- 1163 Wooster, M. J., Roberts, G., Perry, G. L. W., & Kaufman, Y. J. (2005). Retrieval of biomass
1164 combustion rates and totals from fire radiative power observations: FRP derivation and
1165 calibration relationships between biomass consumption and fire radiative energy release.
1166 *Journal of Geophysical Research: Atmospheres*, 110(24), 1–24.
1167 <https://doi.org/10.1029/2005JD006318>
- 1168 Xu, W., Wooster, M. J., He, J., & Zhang, T. (2021). Improvements in high-temporal resolution
1169 active fire detection and FRP retrieval over the Americas using GOES-16 ABI with the
1170 geostationary Fire Thermal Anomaly (FTA) algorithm. *Science of Remote Sensing*, 3,
1171 100016. <https://doi.org/10.1016/j.srs.2021.100016>
- 1172 Yokelson, R.J., Griffith, D.W. & Ward, D.E., 1996. Open-path Fourier transform infrared studies
1173 of large-scale laboratory biomass fires. *Journal of Geophysical Research: Atmospheres*,
1174 101(D15), 21067-21080. <https://doi.org/10.1029/96JD01800>
- 1175 Yokelson, Robert J., Susott, R., Ward, D. E., Reardon, J., & Griffith, D. W. T. (1997). Emissions
1176 from smoldering combustion of biomass measured by open-path Fourier transform infrared
1177 spectroscopy. *Journal of Geophysical Research: Atmospheres*, 102(15), 18865–18877.
1178 <https://doi.org/10.1029/97jd00852>

1179 Yokelson, R. J., Goode, J. G., Ward, D. E., Susott, R. A., Babbitt, R. E., Wade, D. D., Bertschi,
1180 I., Griffith, D. W. & Hao, W. M. (1999). Emissions of formaldehyde, acetic acid, methanol,
1181 and other trace gases from biomass fires in North Carolina measured by airborne Fourier
1182 transform infrared spectroscopy. *Journal of Geophysical Research: Atmospheres*,
1183 *104*(D23), 30109–30125. <https://doi.org/10.1029/1999JD900817>

1184 Yokelson, R. J., Burling, I. R., Gilman, J. B., Warneke, C., Stockwell, C. E., Gouw, J. D., Akagi,
1185 S. K., Urbanski, S. P., Veres, P., Roberts, J. M. & Kuster, W. C. (2013). Coupling field and
1186 laboratory measurements to estimate the emission factors of identified and unidentified
1187 trace gases for prescribed fires. *Atmospheric Chemistry and Physics*, *13*(1), 89–116.
1188 <https://doi.org/10.5194/acp-13-89-2013>

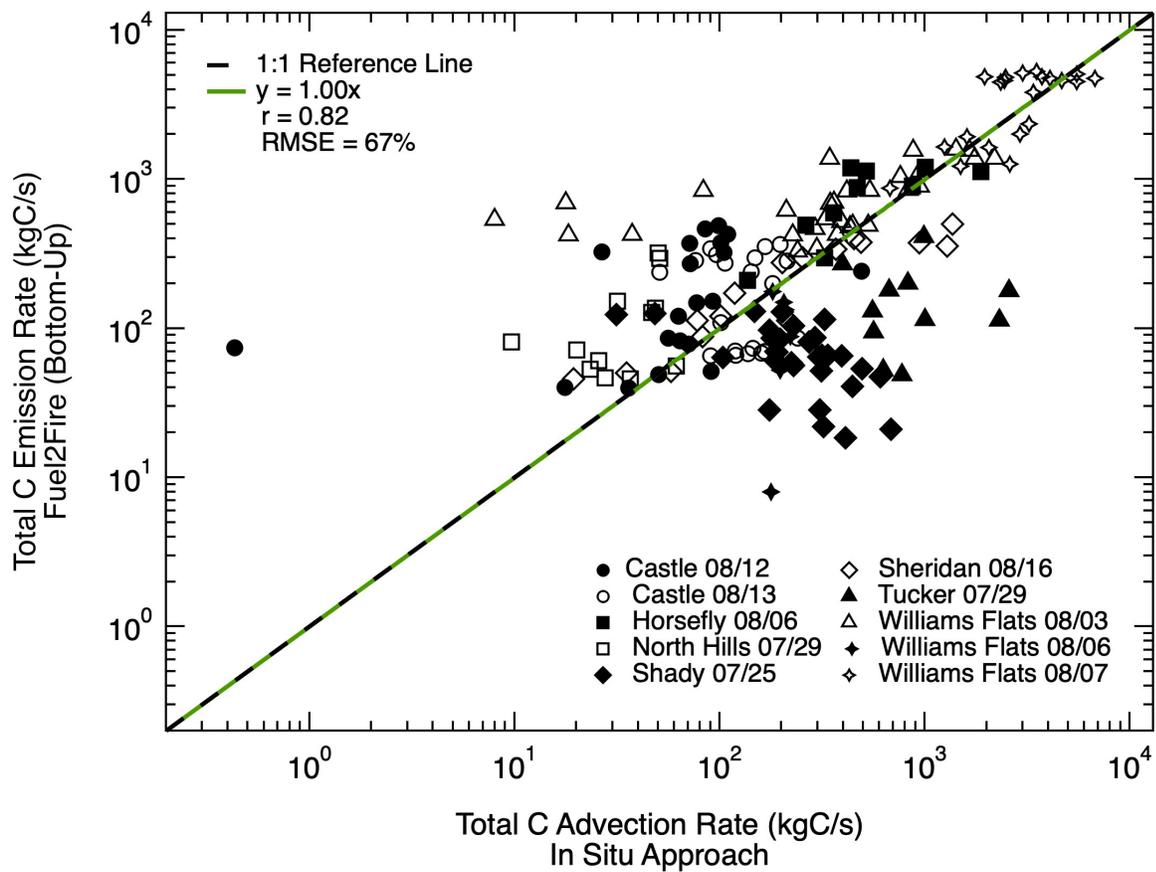
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1190 **Figures and Tables**
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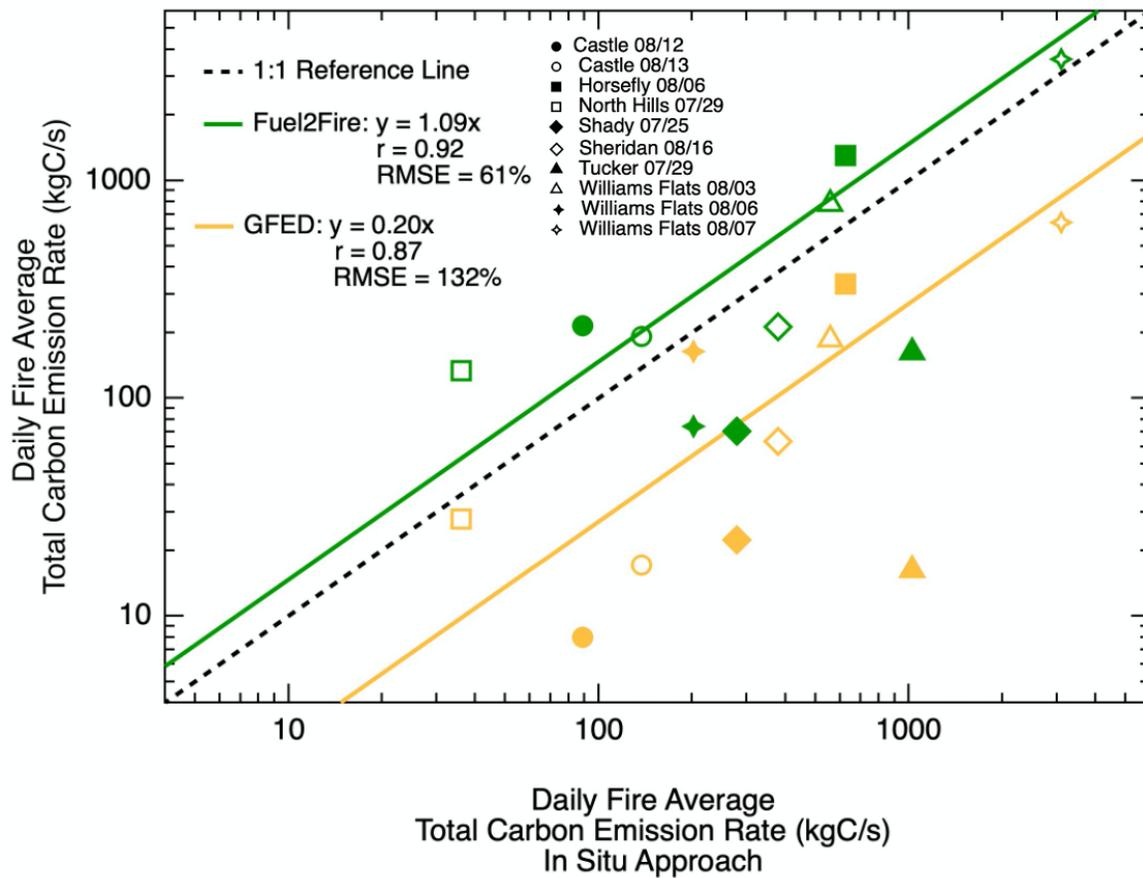
Figure 1. Conceptual image of a typical wildland fire and smoke plume observed during FIREX-AQ as well as the observational platforms and analysis approaches. The DC-8 flight track is given in red and colored by in situ particle concentrations for the cross-sectional legs. As described in the text, the DC-8 initially completes a longitudinal run where the nadir HSRL measurement provides the full smoke curtain below the aircraft, which is then followed by a series of successively downwind flight legs where the nadir- and zenith-pointing HSRL curtains are used to contextualize the cross-sectional, in situ measurements. Image credit: NASA / Tim Marvel.



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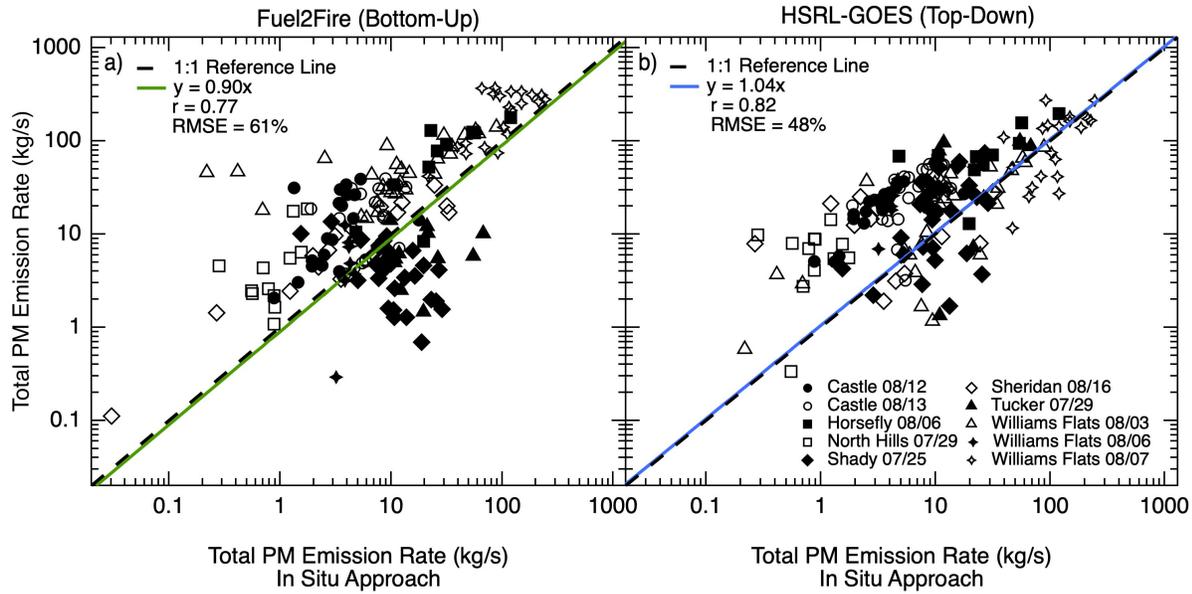
1203 **Figure 2.** Relationship between total carbon emission rates (E_C) from the high resolution bottom-
 1204 up approach, Fuel2Fire, and the in situ approach. Different markers correspond to specific fires
 1205 and repeated markers correspond to different transects of the same fire. The green line shows the
 1206 fit between E_C using a reduced major axis regression with a forced zero intercept. The dashed
 1207 black line shows a perfect 1:1 relationship for reference. The slope for the linear fit, Pearson's
 1208 correlation coefficient (r), and root mean square error (RMSE) is given in the legend.

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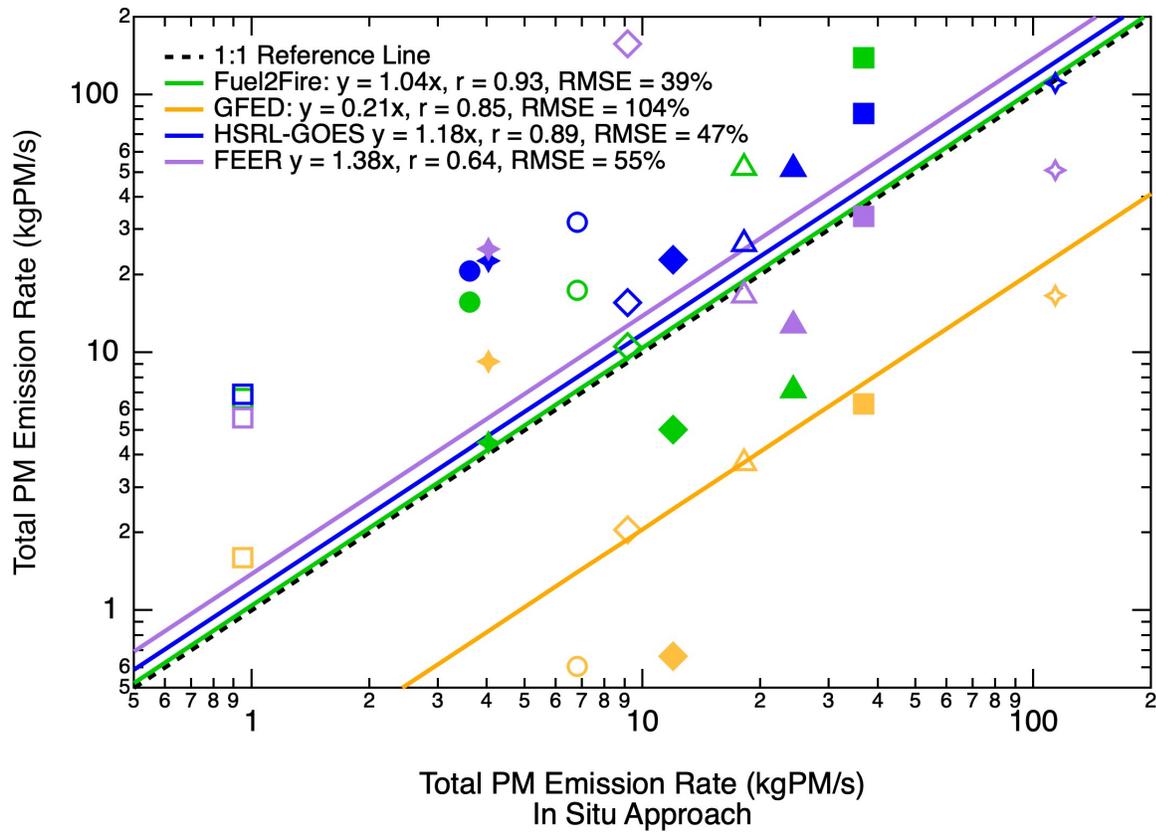
1211 **Figure 3.** Relationship between daily fire average total carbon emission rates (E_C) from
 1212 Fuel2Fire and GFED versus the in situ measurement based approach. Different markers
 1213 correspond to specific fires. The green line shows the fit between Fuel2Fire E_C estimates versus
 1214 the in situ approach using a reduced major axis regression with a forced zero intercept. The
 1215 yellow line shows the fit between GFED E_C estimates versus the in situ approach. The dashed
 1216 black line shows a perfect 1:1 relationship for reference. The slope for the linear fit, Pearson's
 1217 correlation coefficient (r), and root mean square error (RMSE) is given in the legend.



1218

1219 **Figure 4.** Relationship between total PM emission rates (E_{PM}) derived from the high-resolution
 1220 bottom-up approach (Fuel2Fire) versus in situ shown in panel a, and the same relationship
 1221 between the high resolution top-down aircraft approach (HSRL-GOES) and the in situ approach
 1222 shown in panel b. The green line shows the reduced major axis regression with a forced zero
 1223 intercept for Fuel2Fire E_{PM} estimates versus in situ, and the blue line shows the fit for the HSRL-
 1224 GOES E_{PM} estimates versus in situ. Legend gives the slope for the linear fit, Pearson's
 1225 correlation coefficient (r), and root mean square error (RMSE).

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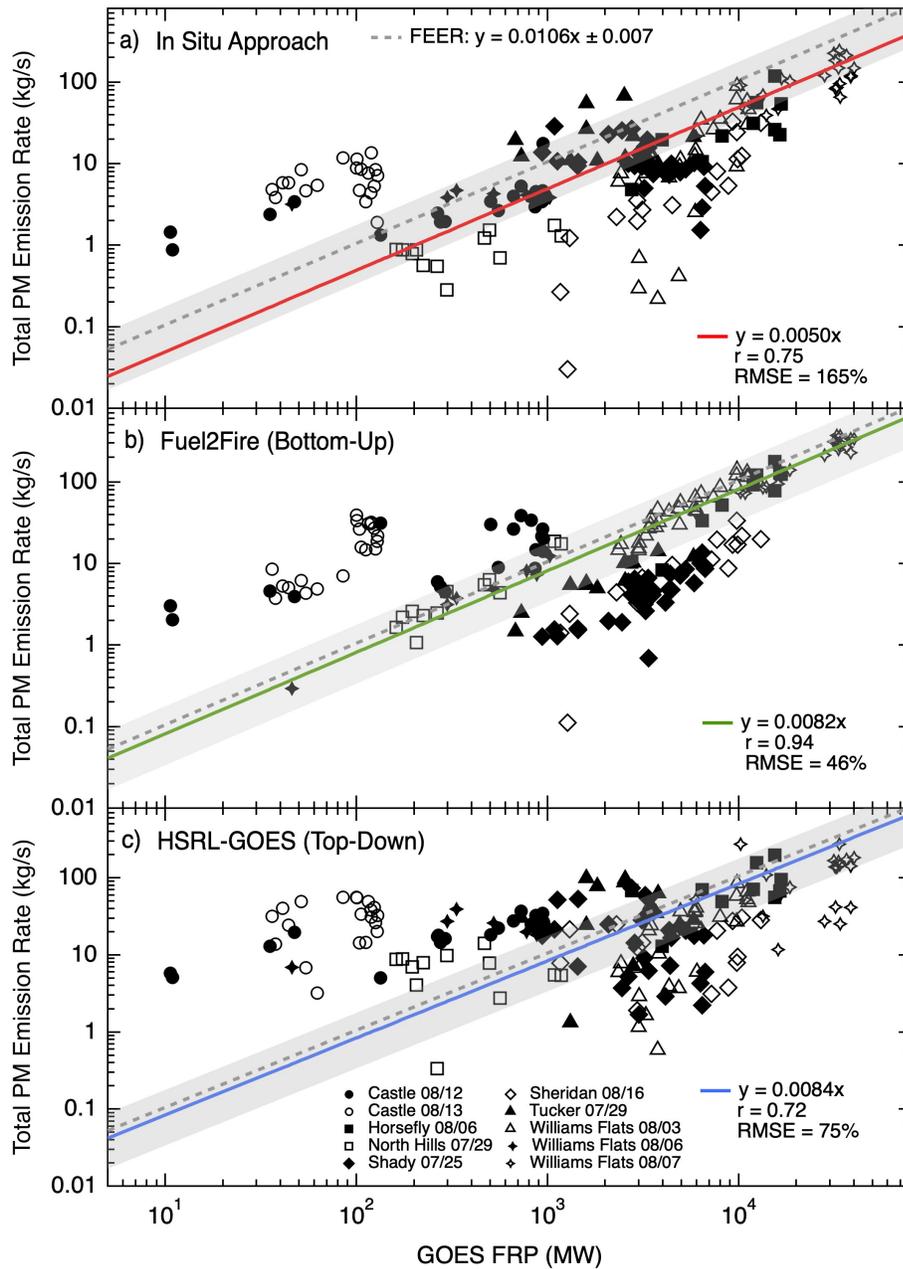


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1228 **Figure 5.** Daily fire average PM emission rates (E_{PM}) from Fuel2Fire, HSRL-GOES, GFED, and
 1229 FEER compared to estimates from the in situ approach. Green markers represent estimates from
 1230 Fuel2Fire and the green line represents the reduced major axis regression with a forced zero
 1231 intercept between Fuel2Fire estimates and in situ estimates. Blue markers and line represent
 1232 HSRL-GOES estimates and regression. Purple markers and line represent FEER estimates and
 1233 regression. Orange markers and line represent GFED estimates and regression. The slope for the
 1234 linear fit, Pearson's correlation coefficient (r), and root mean square error (RMSE) is given in the
 1235 legend.

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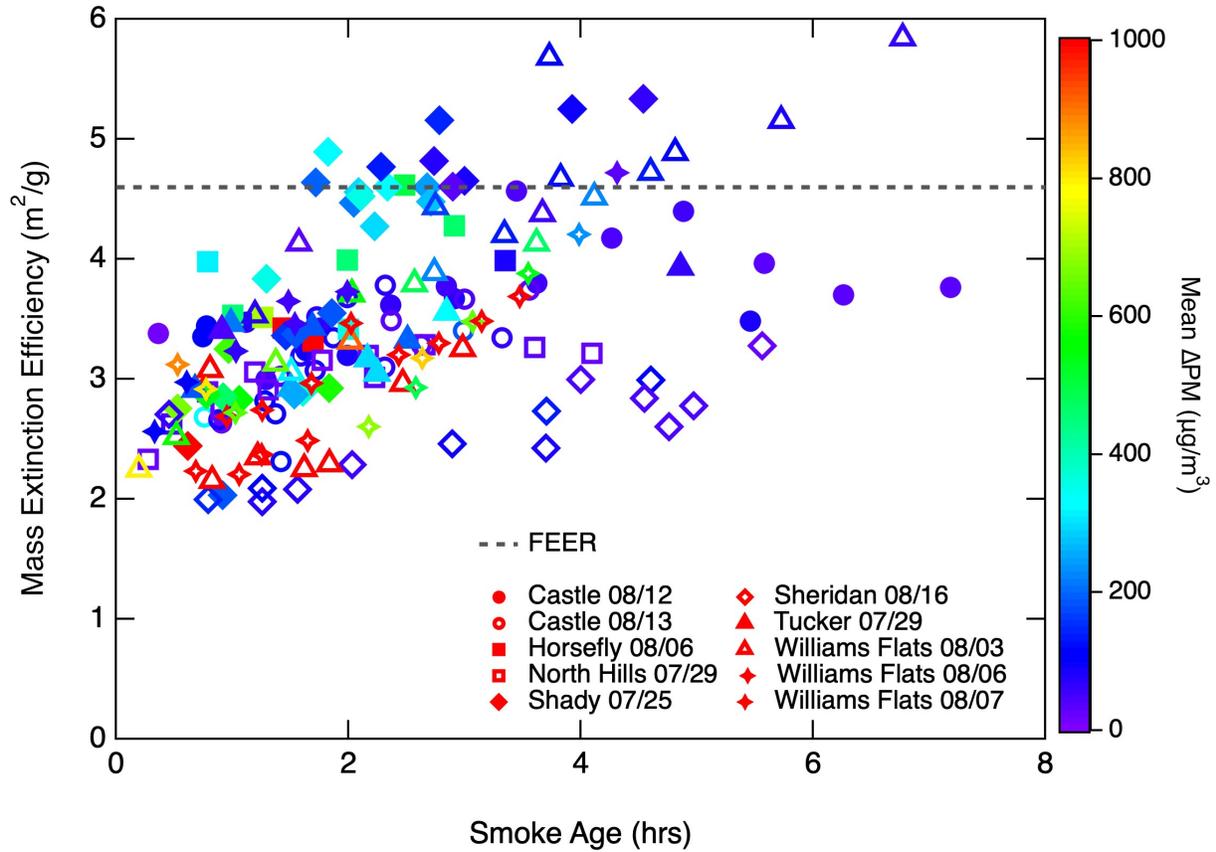
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1239 **Figure 6.** Relationship between GOES FRP and total PM emission rates (E_{PM}) derived from the
 1240 in situ approach (panel a) and the same relationship for Fuel2Fire (panel b) and HSRL-GOES
 1241 (panel c). The red line shows the fit to a reduced major axis regression with a forced zero
 1242 intercept for the GOES FRP versus in situ comparison, the green line shows the fit for Fuel2Fire,
 1243 and the blue line shows the fit for HSRL-GOES. The slope of each regression is equal to the
 1244 smoke emission coefficient (C_e). The dashed grey line is the C_e derived from FEER and the grey
 1245 shading represents the corresponding uncertainty range. Legend gives the slope for the linear fit,
 1246 correlation coefficient (r), and root mean square error (RMSE %).

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1249 **Figure 7.** Mass extinction efficiency (MEE) versus smoke age per transect for each fire. Markers
 1250 are colored as a function of transect mean excess PM concentration. The constant MEE assumed
 1251 by FEER is shown as the dashed black line for reference.

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1253

Inventory or Approach	Style	Spatial Range	Temporal Resolution	Eqns.	Input Variables	Output Variables
GFED4.1s	Bottom-up	Global	Daily	1	BA, FL, CC, F _C	E _C
				2	$\hat{\mathbf{E}}_{\text{C}_{\text{GFED}}}$, EF _{PM} , F _C , $\Delta\mathbf{X}_{\text{GFED}}$	E _{PM}
FEERv1.0	Top-down	Global	Daily	3	C _c (MODIS), FRP (MODIS)	E _{PM}
				4	$\hat{\mathbf{E}}_{\text{PM}_{\text{FEER}}}$, $\Delta\mathbf{X}_{\text{FEER}}$	E _{PM}
In Situ	In situ	Western US (FIREX-AQ)	Sub-plume timescale (per aircraft transect)	5	CO ₂ , CO, CH ₄ , OC, BC, PM, H, WS, GS	E _C , E _{PM}
				6	$\Delta\mathbf{z}$, $\boldsymbol{\beta}$	H
Fuel2Fire	Bottom-up	Western US (FIREX-AQ)	Sub-plume timescale (per aircraft transect)	1	BA, FL, CC, F _C	E _C
				7	E _C , EF _{PM} , F _C	E _{PM}
HSRL-GOES	Top-down	Western US (FIREX-AQ)	Sub-plume timescale (per aircraft transect)	3	C _c (Aircraft-GOES), FRP (GOES)	E _{PM}
				8	WS, GS, MEE, FRP (GOES), AOT	C _c
				9	$\boldsymbol{\alpha}_t$, $\Delta\mathbf{z}$	AOT

1254 **Table 1.** Summary of approaches used to calculate fire carbon and PM emission rates. Note
 1255 GFED4.1s also provides data at a 3hr temporal resolution, but we use only the daily product.

1256

Fire Name	Date Flown	Fuel2Fire E_{PM}			HSRL-GOES E_{PM}		
		m	r	RMSE	m	r	RMSE
Shady	07/25	0.13	0.44	13%	1.69	0.53	67%
North Hills	07/29	1.69	0.45	58%	6.27	0.55	30%
Tucker	07/29	0.10	0.61	39%	1.59	0.66	107%
Williams Flats	08/03	1.13	0.89	15%	1.07	0.84	149%
Williams Flats	08/06	0.59	0.07	116%	5.76	0.33	37%
Horsefly	08/06	1.70	0.63	627%	1.92	0.89	15%
Williams Flats	08/07	0.88	0.63	45%	0.94	0.69	87%
Castle	08/12	1.10	0.56	29%	3.54	0.73	18%
Castle	08/13	1.15	0.53	232%	4.48	0.71	232%
Sheridan	08/16	0.41	0.77	1529%	0.93	0.69	276%

1257 **Table 2.** Reduced major axis regression slope (m), Pearson's correlation coefficient (r), and root
1258 mean square error (RMSE) for PM emission rates (E_{PM}) from Fuel2Fire and HSRL-GOES versus
1259 the in situ based approach per fire. Fire name is given in the far left panel, followed by date
1260 flown.

1261

Fire Name	Date Flown	In Situ			Fuel2Fire			HSRL-GOES		
		m	r	RMSE	m	r	RMSE	m	r	RMSE
Shady	07/25	0.0022	0.53	85%	0.001	0.80	148%	0.012	0.44	78%
North Hills	07/29	0.0011	0.77	57%	0.014	0.95	46%	0.015	0.61	40%
Tucker	07/29	0.0076	0.49	74%	0.003	0.89	55%	0.033	0.60	63%
Williams Flats	08/03	0.0039	0.69	934%	0.010	0.91	71%	0.011	0.69	139%
Williams Flats	08/06	0.0039	0.70	6%	0.010	0.96	491%	0.057	0.63	69%
Horsefly	08/06	0.0012	0.61	292%	0.008	0.84	311%	0.012	0.69	118%
Williams Flats	08/07	0.0040	0.67	21%	0.009	0.90	1%	0.008	0.45	18%
Castle	08/12	0.0060	0.58	4531%	0.027	0.68	209%	0.060	0.86	111%
Castle	08/13	0.0646	0.61	57%	0.204	0.65	147%	0.555	0.55	81%
Sheridan	08/16	0.0017	0.68	3154%	0.002	0.84	1199%	0.005	0.66	274%

1262 **Table 3.** Reduced major axis regression slope (m), Pearson's correlation coefficient (r), and root
1263 mean square error (RMSE) for GOES FRP versus total PM emission rates (E_{PM}) for the in situ
1264 approach, Fuel2Fire, and HSRL-GOES per individual fire. The slope is equal to the smoke
1265 emission coefficient (C_e).

1266