

1 Estimation of Fire-induced CO Plume Age from NAST-I During the 2 FIREX-AQ Field Campaign

3
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7 **Abstract.** Ultra-spectrally resolved infrared measurements from aircraft and space-based observations contain
8 information about tropospheric carbon monoxide (CO) and ozone (O₃), as well as other trace species. A methodology
9 for retrieving these tropospheric trace species from such remotely sensed spectral data has been developed and
10 validated for the National Airborne Sounder Testbed-Interferometer (NAST-I). The Fire Influence on Regional to
11 Global Environments and Air Quality (FIREX-AQ) field campaign was conducted during August 2019 to investigate
12 the impact of wildfire and biomass smoke on air quality and weather in the continental United States. NAST-I CO
13 and O₃ measurements from the recent FIREX-AQ field campaign are presented herein and used to estimate wildfire
14 plume age. Results show enhanced levels of CO in the evolving plume as it is transported away from the fire ground
15 site, and its plume age associated with the plume distance in both vertical and horizontal directions from the wildfire
16 location. These results are enabled by the moderate-vertical and high-horizontal resolution obtained from the NAST-I
17 IR spectrometer onboard NASA ER-2 aircraft. This study advances our knowledge of fire-induced plumes with
18 their evolution and age characterized in 3-dimensional space using information from NAST-I retrieved CO and O₃
19 and relative changes in their concentrations.

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21 **Keywords:** Plume age, ozone, carbon monoxide, wildfires, infrared measurements, remote sensing.
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25 1 Introduction

26 In recent decades, wildfires have gained more of our attention as their number of occurrences,
27 sizes, and intensities have significantly increased, likely due to climate change, or dryer and hotter
28 conditions. Wildfire-induced pollutants and smoke (i.e., poor air quality) pose great risks to human
29 health although wildfires can be an important natural event in many ecosystems. Chemistry and
30 composition of smoke from wildfires are being studied to improve our understanding of the
31 relationship between combustion and air quality, weather, and climate and the ability to forecast.¹

32 Carbon monoxide (CO) is one of the major pollutants due to combustion.² The significance
33 of CO in atmospheric chemistry was recognized long ago when a photo-chemically driven chain
34 reaction was recognized linking the tropospheric cycles of CO, methane (CH₄), and ozone (O₃)

35 with those of the hydroxyl radical (OH) and hydroperoxyl radical (HO₂).³ O₃ as another major
36 pollutant can also cause several health problems to human beings.⁴ O₃ plays a significant role in
37 tropospheric chemistry; and details on ozone production from wildfires can be found from other
38 studies.^{5,6} Previous studies and observations suggested some degree of O₃ production as wildfires
39 generate emissions of O₃ precursors, such as NO_x (NO+NO₂) emitted from wildfires thus
40 increasing PAN (peroxyacetyl nitrate) as well.⁶ O₃ production decreases quickly downwind of
41 combustion.⁷ However, O₃ production can be complicated and depend upon many factors, e.g.,
42 aerosols in a biomass plume from a wildfire reduce the photolysis rates of NO₂ and O₃. The impact
43 of lower photolysis rates on O₃ production is not clear. Reducing the photolysis rates can either
44 increase or decrease the net O₃ production by changing both the O₃ production and loss rates.⁶ O₃
45 plume characteristics from wildfires are not as obvious as that for CO plumes because O₃ is not
46 directly produced by wildfires, and it has a short lifetime in comparison to that of CO. However,
47 both CO and O₃ within wildfire plumes are particularly interesting as they are related to the plume
48 age and associated plume evolution and transport. Observations were made of $\Delta O_3/\Delta CO$ due to
49 wildfires for biome and plume age,⁶ indicating a positive relationship between $\Delta O_3/\Delta CO$ and
50 plume age. Tropospheric chemical reactions involving CO extend their influence on air quality
51 and global climate through accumulation of greenhouse gases. CO can be transported a great
52 distance from its original source due to its relatively long lifetime (averaging about 2 months) in
53 the troposphere.

54 The Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ)
55 experiment, during August 2019 is the first joint field campaign conducted by NOAA and NASA
56 addressing wildfire emissions and their impact on air quality and climate. It was dedicated to the
57 sampling and characterization of fires and their impact on air quality and weather from the point

58 of trace species emissions.^{8,9} Ground, airborne, and satellite measurements were made during the
59 FIREX-AQ field phase. The National Airborne Sounder Testbed-Interferometer (NAST-I) is an
60 airborne interferometer sounding system. It provides a highly spatial linear resolution that is equal
61 to 13% of the aircraft altitude at nadir and 13 instrument field-of views (IFOVs) across the aircraft
62 track from 13 scan angles (i.e., 2.6 km IFOV, ~3.4 km apart on the ground from ER-2 altitude of
63 20 km). NAST-I spatially scans and provides high-spectral resolution (0.25 cm^{-1}) measurements
64 within the spectral region of $645\text{--}2700\text{ cm}^{-1}$.¹⁰⁻¹⁵ Here we use measurements from NAST-I on
65 board the NASA ER-2 aircraft to study fire-induced CO plumes. NAST-I continuously covers a
66 space large enough to monitor the wildfire plume from its origination, evolution and transport,
67 providing 3-dimensional (3-d) distributions of geophysical parameters including O_3 and CO with
68 a higher spatial resolution comparing to satellite IR-ultraspectral sensors and thus, benefits our
69 study of wildfire plumes.¹⁶ NAST-I data used in this ~~letter~~ study were collected under clear-sky
70 conditions. Geophysical parameters cannot be retrieved with NAST-I under opaque clouds as
71 infrared measurements are not able to penetrate opaque clouds.¹⁴ Materials presented in this ~~letter~~
72 study are a follow-up to the work of Zhou et al.¹⁶ A brief description of the FIREX-AQ
73 experiment, observations, and wildfire plume age estimation methodology will be given in Section
74 2. Results and discussion are presented in Section 3. Summary and concluding remarks follow in
75 Section 4.

76 **2 Experiment, Observation, and Plume Age Estimation Methodology**

77 FIREX-AQ is a multi-disciplinary effort campaign with multi-agency collaborations to study
78 complex fire systems. Research platforms were heavily instrumented with in-situ and remote-
79 sensing devices to allow exhaustive characterization of gases and aerosols, optical properties, wind
80 fields, fire radiative power and more. Ground-based examinations of fuels and burned areas

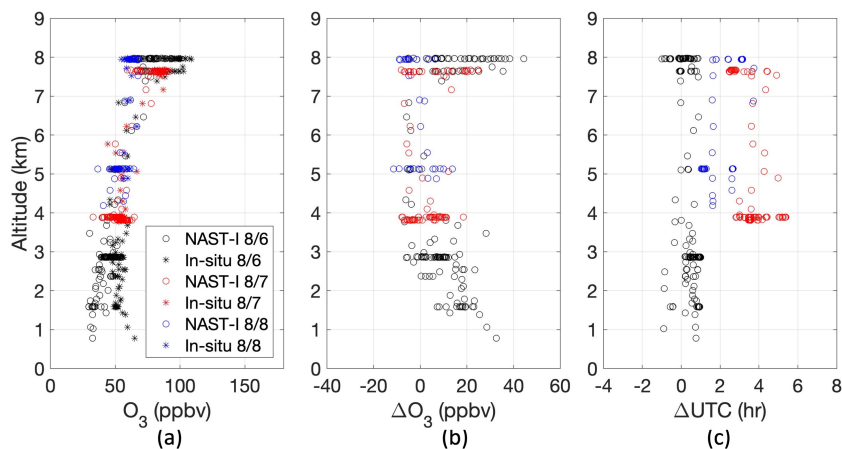
81 permitted clearer connections between atmospheric pollutants and their sources. Modeling efforts
82 were used during the campaign to predict transport, and study emissions and downwind plume
83 transformations.

84 The results presented here are based upon CO and O₃ retrievals from NAST-I measurements.
85 The NAST-I instrument, and its retrieval algorithms are described elsewhere.¹⁰⁻¹⁹ The western
86 portion of the FIREX-AQ campaign domain (August 5–21, 2019) covers 14 large wildfires fueled
87 by grass, woodland, and scrub.²⁰ Fire-induced CO plumes observed by NAST-I during the
88 FIREX-AQ have been analyzed and reported.¹⁶ For this study of wildfire plume age, we have
89 chosen the ER-2 sorties over the William Flats fire and the extended downwind area from August
90 7, 2019, as the ER-2 sorties have ~450 km downwind flight leg segments. The William Flats fire
91 was caused by lightning on August 2, 2019. It covered approximately 100 km² and was located at
92 about 11 km southeast of Keller, Washington. The ER-2 flew from west (-120° longitude) to east
93 (-113° longitude), then back west at a near-constant latitude, passing over the fire location (48.0°
94 latitude, -118.5° longitude) and the extended downwind area to detect fire-induced gas emissions
95 and characterize their subsequent evolution. A large downwind area covered by the ER-2 flight
96 makes an excellent naturally occurring experiment to study fresh and aged plumes.

97 Geophysical parameters such as temperature, moisture, CO and O₃ profiles are retrieved from
98 NAST-I measured spectral radiances. The NAST-I retrieval algorithm was developed, tested,
99 improved, and validated.^{13,14,17} The NAST-I trace gas (CO and O₃) retrieval algorithm was also
100 developed¹⁸ and later improved by implementation of a surface emissivity retrieval.¹⁹ During the
101 FIREX-AQ field campaign, CO retrievals are validated by using in-situ measurements from the
102 NASA DC-8 aircraft.¹⁶ There are two O₃ in-situ sensors flying on the DC-8 aircraft: one is the
103 NOAA Nitrogen Oxides and Ozone (NO_yO₃) 4-channel chemiluminescence instrument²¹ and the

104 other is the Rapid Ozone Experiment (ROZE).²² The nature of O₃ (e.g., its lifetime and
105 photochemical reactions) and its impact on the atmosphere from wildfires is not as obvious as that
106 of CO. O₃ is relatively stable in comparison with fire-induced CO, which makes it a bit easier for
107 inter-comparison between NAST-I remotely sensed and in-situ measured ozone. A few DC-8
108 sorties were spatially coincident with the ER-2 sorties at the same fire locations but, in general,
109 they had lag times of a few hours. There was one exception wherein both spatial and temporal
110 coincidence was achieved between the two aircraft, specifically, on August 6, 2019.¹⁶ Here we
111 use the 60-second merged data from DC-8 measurements available from the FIREX-AQ
112 database²³ to ~~inter~~compare with NAST-I O₃ retrievals. Inter-comparison between ER-2 NAST-I
113 O₃ and DC-8 in-situ O₃ are made in the vicinity of the William Flats fire location from data
114 collected on August 6–8, 2019. Fig. 1a plots the NAST-I O₃ (in black open circles) and in-situ O₃
115 data (the mean of ROZE and NO_yO₃ measurements, in black asterisks) with a coincidence spatial-
116 criteria of $|\Delta(\text{latitude})| < 0.05^\circ$ and $|\Delta(\text{longitude})| < 0.05^\circ$ and temporal-criteria of $|\Delta(\text{UTC})| < 1.0$
117 hr- for August 6, 2019. The NAST-I O₃ profile is interpolated to DC-8 in-situ altitude. Fig. 1b
118 shows the difference between NAST-I and in-situ O₃ plotted in Fig. 1a. Similar data from August
119 7 and 8, 2019, are also plotted (in red and blue symbols, respectively) with a relatively larger
120 temporal-criteria of $|\Delta\text{UTC}|$ as shown in Fig. 1c. Overall, from the data shown in Fig. 1, the mean
121 error (bias) and standard deviation error (STDE) between NAST-I retrievals and in-situ
122 measurements are 7.55 (ppbv) and 10.85 (ppbv), respectively. Reasonable agreement between the
123 ER-2 NAST-I and DC-8 in-situ O₃ measurements is achieved with some differences due to their
124 spatial and temporal mismatches. However, it is noticed that NAST-I O₃ starts to deviate from in-
125 situ measurements at an altitude of ≤ 3 km. This is believed to be a consequence of the expected
126 lower ozone retrieval sensitivity in the NAST-I passive infrared measurements at an altitude region

127 below 3 km caused by a large amount of O_3 from higher altitudes in the lower stratosphere.
 128 However, the uncertainty of ΔO_3 is relatively small compared to that of O_3 itself as the retrieval
 129 uncertainties of the polluted and clean background O_3 regions are similar. From this evaluation,
 130 we believe that NAST-I O_3 retrievals are reasonably good at the altitude of 3 km and above and
 131 can be used together with CO retrievals to estimate the age of wildfire-induced plumes as presented
 132 in the following section.

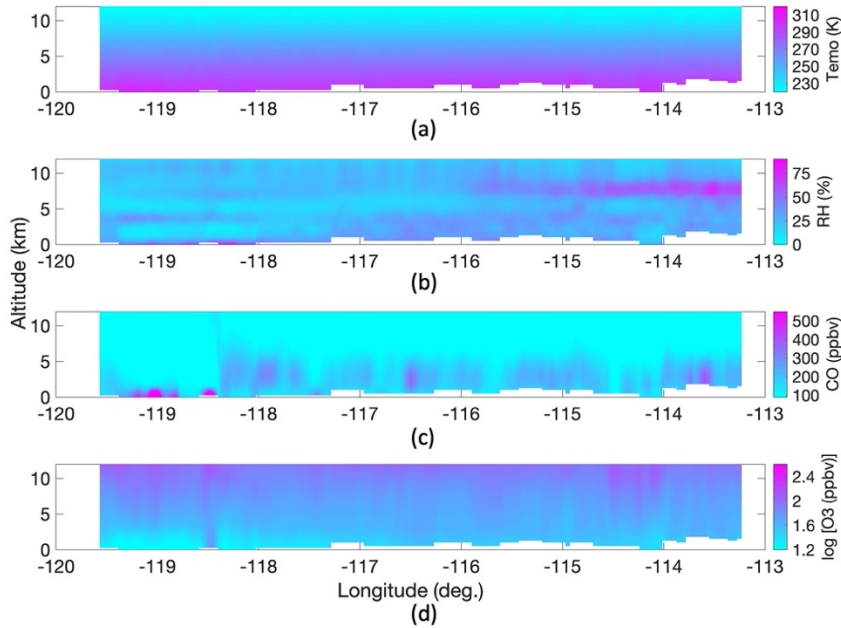


133 **Fig. 1** NAST-I O_3 retrieval evaluation with in-situ measurements near William Flats fire location from August 6–8,
 134 2019, in black, red, and blue, respectively (see text). (a) NAST-I retrieved and In-situ measured O_3 , (b) the
 135 difference between NAST-I and In-situ O_3 , and (c) the UTC difference between NAST-I and In-situ O_3
 136 measurement.
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139 The William Flats fire progression and CO plume evolution during August 6–8 2019 were
 140 observed by multiple airborne sensors including NAST-I.¹⁶ NAST-I CO retrieval uncertainty has
 141 been evaluated with the co-incident In-situ measurements, and found to have a bias of ~ 8 ppbv
 142 and standard deviation of difference (STDE) of ~ 28 ppbv.¹⁶ The NAST-I measurements provide
 143 3-d distribution of temperature, water vapor, CO, and O_3 . For the case study here, the ER-2 sorties
 144 went over the William Flats fire and the extended downwind area twice. NAST-I retrieved cross
 145 sections in nadir view are plotted in Fig. 2 for one sortie (from west to east); the retrievals from
 146 the return sortie are almost identical. Moisture layers are shown in relative humidity (RH) with

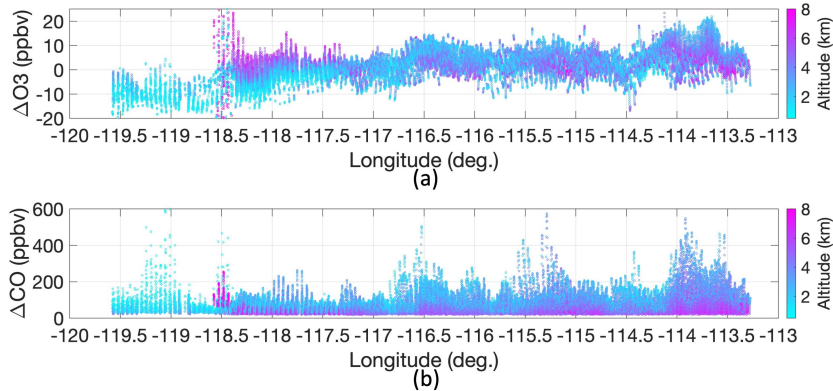
147 less saturation (<100%) indicating clear-sky conditions in the observations. CO plumes in the
148 downwind area are evidently clear. The O₃, plotted in a logarithmic scale, illustrates its small
149 variation in free troposphere from the ground level up and its enhancement over the fire location
150 (-118.5° longitude).

151 Fire-induced plumes contain information on chemical gaseous photochemical reaction and
152 production; it is essential to know the plume's evolution and age to better understand impacts to
153 air quality, weather, and climate. Fire plume age has been widely measured and studied by
154 numerous researchers. Observations of $\Delta O_3/\Delta CO$ due to wildfires by biome and plume age were
155 summarized by Jaffe and Wigder.⁶ Many factors contribute to O₃ production within wildfire
156 plumes. In general, Jaffe and Wigder found a positive relationship between the $\Delta O_3/\Delta CO$ ratio
157 and plume age. Here we assume this relationship is a linear $T = \alpha R + \beta$, where T is plume age in
158 hours and R is $\Delta O_3/\Delta CO$ ratio. α and β are the slope and intercept, respectively. A fitting
159 relationship is obtained from Table 1 of Jaffe and Wigder.⁶ For boreal and temperate regions
160 (BTR), $\alpha=327.2$ and $\beta=89.4$; while for the tropical and subtropical regions (TSR), $\alpha=215.8$ and
161 $\beta=3.1$. The $\Delta O_3/\Delta CO$ ratio increases as the plume ages, and it ages relatively slower in tropical
162 and equatorial regions as more O₃ production expected from more NO_x emissions per unit of fuel
163 consumed.⁶ Based upon this assessment, our plume age estimation methodology relies on
164 $\Delta O_3/\Delta CO$ ratios within the plumes.



165 **Fig. 2** NAST-I retrieval cross sections in nadir view for (a) temperature (K), (b) relative humidity (%), (c) CO
 166 (ppbv), and (d) O₃ (ppbv) in logarithmic scale.
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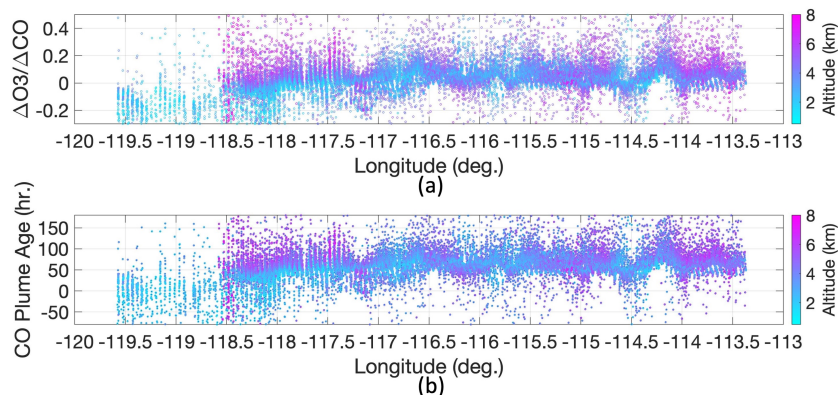
169 Within fire-induced CO plumes, we assume that CO concentrations are greater than 135 ppbv.
 170 Subtracting the background estimates, ΔO_3 and ΔCO are calculated using NAST-I retrieved O₃
 171 and CO. O₃ background is assumed to be the average of its regional climatology which is from a
 172 global climatology database that consists of 15,150 profiles obtained from the extended SeeBor
 173 database of Univ of Wisconsin-Madison^{23,24} and NAST-I retrieval mean, while CO background is
 174 assumed to be its regional climatology. 3-d distributions of ΔO_3 and ΔCO within the plumes
 175 (where CO > 135 ppbv) are plotted in Figs. 3a and 3b, respectively, along the longitude with a
 176 color distribution in altitude. It is noted that elevated O₃ spans at least 26 km near the fire location
 177 (-118.5° longitude) and covers an area of 531 km². It is worth mentioning that ΔO_3 and ΔCO
 178 plotted in Fig. 3 are estimations as O₃ and CO background within the plumes are not precisely
 179 known but made to the best of our knowledge and assumed to be the same throughout the local
 180 region.



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182 **Fig. 3** (a) ΔO_3 and (b) ΔCO distribution within the fire-induced plumes.
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184 **3 Results and Discussion**

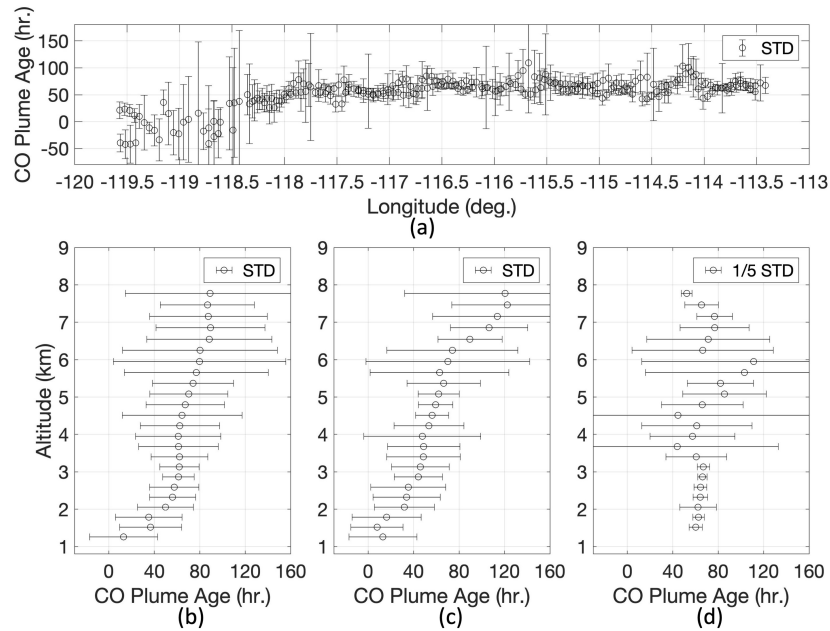
185 The $\Delta O_3/\Delta CO$ ratio is estimated from retrievals using NAST-I flight observations and plotted in
186 Fig. 4a. CO plume age can be simply projected from wildfire $\Delta O_3/\Delta CO$. For the William Flats
187 fire location (48.0° latitude), a linear combination of 55% BTR and 45% TSR coefficients are used
188 to calculate the plume age that would reflect a near fresh plume at the fire location. Estimated
189 plume age is plotted in Fig. 4b. For plumes at a lower altitude (i.e., 4 km and below), their ages
190 increased as the plumes moved away from the fire location downwind and merged with aged
191 plumes (~60–100 hr.) while other aged plumes were sitting at a higher altitude (4 km and above).



192 **Fig. 4** (a) $\Delta O_3/\Delta CO$ distribution along the longitude within the fire-induced plumes, and (b) estimated plume age
193 distribution.
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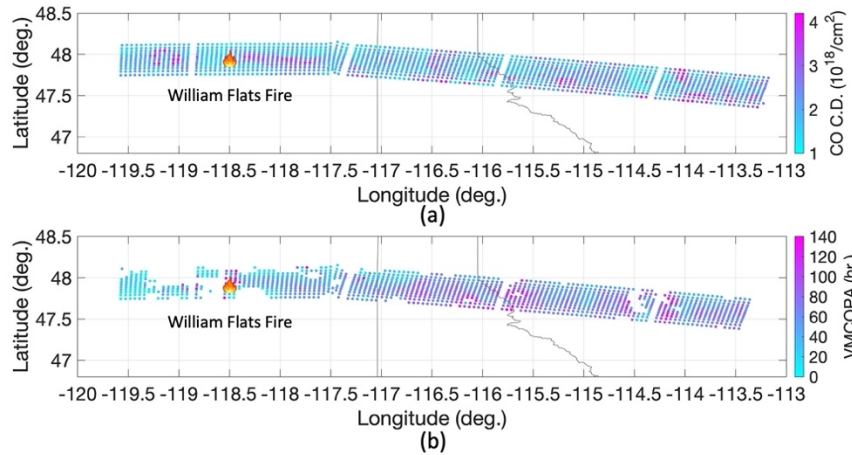
196 Fig. 5a plots the plume age along the longitude with error bars indicating its variation over

197 altitude and longitude. Fresh plumes were observed to be closer to the fire location while aged
198 plumes were in the downwind regions as expected. Figs. 5b-5c plots the mean plume age against
199 the altitude with an error bar indicating its variation over longitude and latitude. The ages depend
200 on the distance (e.g., longitude) from the fire location. Fig. 5b covers all data shown in Fig. 5a;
201 Figs. 5c and 5d have data plotted where longitude is less than and greater than -117° , respectively.
202 Near the fire location, shown in Fig. 5c, fresh plumes are at a lower altitude and plume ages
203 increase as altitude increases. As the plumes move further downwind, as shown in Fig. 5d, fresh
204 and aged plumes were mixed along the altitudes with a near constant mean plume age of ~ 50 hours,
205 but with a large variation (i.e., a large standard deviation) indicated by the error bars (reduced by
206 a factor of 5 for clarity) in the altitude region of $\sim 3.5\text{--}7.0$ km, where a greater mixture of fresh and
207 aged plumes co-exist. In general, as shown in Fig. 4b, fresh plumes were at a lower altitude while
208 aged plumes were found in the upper regions. Lastly, the distributions of enhanced CO column
209 density from the fire plume and the vertical mean CO plume age (VMCOPA) are plotted in Figs.
210 6a and 6b, respectively. It is worth noting that there are two flight tracks (2 legs) back and forth
211 with time evolution on top of each other, only the first leg is plotted in Fig. 6 for clarity. Comparing
212 CO column density and its vertical mean CO plume age, a large amount of relatively fresh CO
213 plumes was near the fire location and aged CO plumes had been transported further downwind
214 and/or upper regions.



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Fig. 5 (a) Plume age distribution along the longitude. Plume age distribution along the altitude: (b) all data shown from (a), (c) data from longitude less than -117.0° (near fire location), and (d) data from longitude greater than -117.0° (further away from fire location).



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Fig. 6 Distribution of (a) CO column density and (b) vertical mean CO plume age (VMCOPA).

223 The William Flats fire started on August 2, 2019, and the oldest plumes could be 4–5 days old.
224 The plume ages could be from mixed plumes (or effective plumes) as indicated in Figs 4-6;
225 therefore, in the area farthest away from the fire location, they could be somewhat less than 5 days
226 old in our estimations. Nevertheless, the plume age distributions, both horizontally and vertically
227 (shown in Figs. 5-6), make sense for the way plumes are transported and aged. The error or

228 uncertainty of plume age derived here is largely due to (1) O₃ and CO retrieval error especially at
229 lower altitudes (3 km and below) as retrieval sensitivity decreased, (2) the uncertainty from
230 estimated CO and O₃ background within the plume, (3) the limited data available for deriving the
231 relationship between the plume age and ΔO_3 to ΔCO ratio, and a large data scattering and
232 uncertainty in the dataset⁶, (4) a linear fitting relationship between the plume age and $\Delta O_3/\Delta CO$
233 ratio may not be the best representation, and (5) mixed (or effective) plumes being assessed,
234 possibly contain contributions from multiple fire sources, especially in locations further away from
235 the primary fire location.

236 Currently these error sources are not completely quantified, therefore, the plume age presented
237 in this ~~letter~~ study is only an estimation. Regardless, however, it is critically important even though
238 it may lack some quantitative accuracy since it nicely demonstrates the ability to characterize
239 wildfire plumes and estimate their age from the perspective of an advanced ultraspectral infrared
240 remote sounder. CO and O₃ retrievals are the mean over the IFOV with a vertical column
241 resolution. The plume we dealt with is an effective plume or a volume mean of mixed plumes.
242 The plume age presented herein is also an effective mean plume age in the NAST-I sensor IFOV,
243 which can be different from in-situ measurements.

244 It is worth mentioning that the North Hills fire, started on July 26, 2019, was located at about
245 5 km northwest of Lake Helena, Montana (46.8° latitude, -112.0° longitude). It covered
246 approximately 20 km². CO plumes from the North Hills fire are unlikely but could contribute to
247 the data presented here at the locations near the western (possibly upwind) areas of the North Hills
248 fire location.

249 **4 Summary and Concluding Remarks**

250 The FIREX-AQ field campaign with multiple aircraft in-situ and remotely sensed observations

251 provides characterization of distributions of chemical species induced by wildfire emissions and
252 subsequent evolution. This unique dataset is very much desirable in validating NAST-I O₃ and
253 CO retrievals and illustrates the benefits of such data for wildfire characterization. The Wildfire
254 case of Williams Flats from the FIREX-AQ experiment reported herein is used to demonstrate a
255 fire-induced plume age estimation approach. Several major summary items and conclusions can
256 be obtained from this work. (1) NAST-I remotely sensed O₃ is evaluated by favorable inter-
257 comparisons with the in-situ O₃ measurements which show a positive agreement (shown in Fig.
258 1). (2) Small but significant enhanced O₃ production near William Flats wildfire location is
259 observed by NAST-I. (3) O₃ and CO productions impacted by the wildfire are estimated within
260 the fire-induced plume. (4) Plume age is estimated using NAST-I observed $\Delta O_3/\Delta CO$ ratios and
261 a linear fitting relationship from previous observations of wildfire $\Delta O_3/\Delta CO$ ratios by biome and
262 plume age.⁶ (5) Plume age distribution both horizontally and vertically indicates how the plume
263 ages as it moves away from the fire location (for William Flats Fire case).

264 It was reported earlier that first-of-a-kind wildfire-induced plume measurements were obtained
265 by the NAST-I ultraspectral remote sensor on board the ER-2 suborbital aircraft, which has shown
266 the intensity and size of wildfire plumes in a high-spatial-resolution of 2.6 km. Now, in the present
267 study, the plume age estimation in a 3-d high-spatial-resolution adds critical temporal information
268 of the fire-induced plume, demonstrating the capability of an ultraspectral remote sensor like
269 NAST-I with a higher spectral and spatial resolution to monitor CO and O₃ and its advantage of
270 giving broader spatial and temporal assessment by rapidly covering a large field of observation.
271 This work demonstrates 3-d plume age estimation and advances our measurement ability to
272 observe fire-induced plumes and characterize their evolution and age.

273 NAST-I was successfully operated during all ER-2 flights of the FIREX-AQ experiment (a

274 total of 11 flights and 50+ hours of science data collected). NAST-I retrievals (e.g., atmospheric
275 temperature, relative humidity, CO, and O₃ profiles, also surface skin temperature), together with
276 experiment data from other satellite/aircraft/ground measurements and analysis from the
277 FIREX-AQ campaign are available²⁵³ for the science community to study wildfire-related topics
278 as described by the overarching objective of FIREX-AQ experiment⁸ and beyond.

279

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360 **Caption List**

361 **Fig. 1** NAST-I O₃ retrieval evaluation with in-situ measurements near William Flats fire location
362 from August 6–8, 2019, in black, red, and blue, respectively (see text). (a) NAST-I retrieved and

363 In-situ measured O₃, (b) the difference between NAST-I and In-situ O₃, and (c) the UTC difference
364 between NAST-I and In-situ O₃ measurement.

365 **Fig. 2** NAST-I retrieval cross sections in nadir view for (a) temperature (K), (b) relative humidity
366 (%), (c) CO (ppbv), and (d) O₃ (ppbv) in logarithmic scale.

367 **Fig. 3** (a) ΔO_3 and (b) ΔCO distribution within the fire-induced plumes.

368 **Fig. 4** (a) $\Delta O_3/\Delta CO$ distribution along the longitude within the fire-induced plumes, and (b)
369 estimated plume age distribution.

370 **Fig. 5** (a) Plume age distribution along the longitude. Plume age distribution along the altitude:
371 (b) all data shown from (a), (c) data from longitude less than -117.0° (near fire location), and (d)
372 data from longitude greater than -117.0° (further away from fire location).

373 **Fig. 6** Distribution of (a) CO column density and (b) vertical mean CO plume age (VMCOPA).

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375 *Authors Biographies*

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