



1 **Tracking organic compounds in smoke plumes using infrared satellite-based measurements**

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23

24 **Abstract**

25 We apply new measurements of methanol, ethene, ethyne, and HCN from the Cross-track  
26 Infrared Sounder (CrIS) to explore the quantitative use of satellite-based thermal infrared (IR)  
27 observations for fire studies. We focus analysis on the western U.S. during the 2018-2019  
28 timeframes of two fire-focused aircraft campaigns, and use the GEOS-Chem model to guide  
29 interpretation. The CrIS data reveal large in-smoke enhancements and species:species  
30 correlations for targeted volatile organic compounds (VOCs), especially during the more active



31 2018 fire year. Spectral enhancements are strongest for methanol and ethene. For VOCs with  
32 similar vertical sensitivities the in-smoke correlations are height-independent and can be  
33 converted to column enhancement ratios without plume altitude information. For VOCs with  
34 dissimilar vertical sensitivities, spectral index correlations change coherently with altitude and  
35 may constrain injection or plume height changes. The mean ( $\pm \sigma$ ) ethene:methanol ratio  
36 measured by CrIS across an ensemble of plumes ( $0.64 \pm 0.24$  mol/mol) matches bottom-up  
37 emission ratios ( $0.63 \pm 0.08$  mol/mol), but satellite-based and aircraft data both reveal greater  
38 variability than is predicted by GEOS-Chem. We propose that fire pyrolysis conditions are one  
39 driver of this variability and use in-situ data to show that near-field ethene:methanol ratios track  
40 pyrolysis conditions and hence inform the abundance of other emitted VOCs. Finally, we apply  
41 CrIS ethene:methanol ratios to estimate the high-temperature pyrolysis fraction for the same  
42 plume ensemble; the resulting fraction correlates with fire radiative power in a manner not well-  
43 captured by models.

44

## 45 1. Introduction

46 Fire-emitted volatile organic compounds (VOCs) drive formation of ozone and secondary  
47 organic aerosols (SOA) (e.g.; Ahern et al., 2019; Alvarado et al, 2015; Alvarado and Prinn,  
48 2009; Bourgeois et al., 2021; Brey and Fischer, 2016; Hatch et al., 2015; He et al., 2024; Jin et  
49 al., 2026; McClure and Jaffe, 2018; Palmo et al., 2025; Xu et al., 2021), with adverse effects on  
50 human and ecosystem health (Cascio et al., 2018; Reid et al., 2016; Fischer et al., 2018; Jaffe et  
51 al., 2008; O'Dell et al., 2021). These effects are expected to intensify in many regions as wildfire  
52 frequency, burned area, and season length increase (Feng et al., 2025; Jones et al., 2022; Brey et  
53 al., 2020). In-situ VOC observations in smoke plumes are sparse and typically capture only a  
54 subset of ambient burning conditions, compromising efforts to diagnose fire emissions and track  
55 the subsequent plume chemistry. Satellite-based VOC measurements from the Cross-track  
56 Infrared Sounder (CrIS; Wells et al., 2025) offer new constraints to help address this gap. Here  
57 we analyze CrIS observations of methanol, ethene, ethyne, and hydrogen cyanide in smoke  
58 plumes across the western U.S. during 2018 and 2019 to assess the information these data can  
59 provide on fire emission processes.

60



61 The amount and speciation of emitted VOCs are governed by the physical and chemical  
62 processes that occur during combustion and by the type of fuel being burned (Crutzen &  
63 Andreae, 1990; Lobert & Warnatz, 1993; Yokelson et al., 1996, 1997; Andreae & Merlet, 2001;  
64 Andreae, 2019; Akagi et al., 2011; Sekimoto et al., 2018, 2023). Relative emission rates for  
65 VOCs containing only carbon, hydrogen, and oxygen depend mainly on combustion conditions;  
66 those for VOCs containing nitrogen, sulfur, or halogens also vary strongly with the fuel  
67 concentration of those elements (Andreae & Merlet, 2001; Roberts et al., 2020). In both cases,  
68 the VOC emissions arise predominantly from pyrolysis of the long-chain polymers contained in  
69 biomass (Lobert and Warnatz, 1993; Yokelson et al., 1996, 1997). Pyrolysis happens  
70 concurrently with the processes of drying, flaming combustion, and smoldering; its contribution  
71 changes over the lifetime of a fire and affects the evolving VOC speciation. For example,  
72 Sekimoto et al. (2018, 2023) find based on a factor analysis of data for the western US that ~85%  
73 of the variability in pyrogenic VOC speciation can be explained by the relative importance of  
74 low- vs. high-temperature pyrolysis. If broadly applicable, this relationship creates an  
75 opportunity for more robust parameterizations of fire emissions.

76 Satellite-based thermal infrared (IR) observations offer valuable chemical information for  
77 understanding fire processes, testing emission estimates, and mapping smoke plume evolution.  
78 Such measurements enable detection of multiple wildfire-relevant species, and unlike  
79 measurements in the ultraviolet and visible (UV/Vis), they are minimally impacted by the fine  
80 aerosols present in smoke. In particular, Wells et al. (2025) have recently developed retrievals  
81 for methanol (CH<sub>3</sub>OH), ethene (C<sub>2</sub>H<sub>4</sub>), hydrogen cyanide (HCN), and ethyne (C<sub>2</sub>H<sub>2</sub>) using  
82 space-borne radiance observations from CrIS. Methanol and ethene are among the top 10  
83 wildfire-emitted non-methane VOCs, with ethyne and HCN also prominent among the top 15  
84 (Permar et al., 2021). HCN is a widely-used fire tracer because it is among the most abundant N-  
85 containing pyrogenic emissions, with few additional sources (Andreae & Merlet, 2001).  
86 Enhancements of all four species have been detected in wildfire smoke by satellite-borne IR  
87 instruments (Clarisse et al., 2011; Alvarado et al., 2011; De Longeville et al., 2021; Wells et al.,  
88 2025), but the capabilities and challenges in using this space-based data for quantitative fire  
89 analyses have not yet been examined in detail.

90



91 In this study, we use thermal IR observations from CrIS to map the distributions of methanol,  
92 ethene, ethyne, and HCN across the western U.S. during the summers of 2018 and 2019. For  
93 each species, we quantify the importance of wildfire smoke in terms of its contribution to the  
94 total signals measured by CrIS. We combine radiative transfer modeling with plume-specific  
95 analyses to explore the impacts of smoke altitude on thermal IR detection sensitivity, and show  
96 that by combining VOCs with similar vertical sensitivities we can quantify species:species  
97 enhancement ratios without requiring any plume height information. We then leverage these  
98 pyrogenic ratios to explore emission variability across individual fires and to assess processes  
99 controlling that variability. Our results provide a foundation to support more widespread use of  
100 thermal IR data in fire science applications.

101

## 102 **2. Methods**

### 103 **2.1 CrIS trace gas retrievals**

104 The CrIS instruments are a set of identical nadir-viewing Fourier Transform spectrometers flying  
105 on three sun-synchronous polar-orbiting satellites with nadir overpass times at ~01:30 and  
106 ~13:30 local time (LT): Suomi-NPP (launched 10/2011), JPSS-1/NOAA-20 (launched 11/2017),  
107 and JPSS-2/NOAA-21 (launched 11/2022). CrIS measures thermal IR radiances at high spectral  
108 resolution ( $0.625\text{ cm}^{-1}$ ) with low noise ( $\sim 0.04\text{ K}$  at  $900\text{ cm}^{-1}$  and  $280\text{ K}$ ) (Zavyalov et al., 2013)  
109 and has a 14 km nadir footprint. Analyses here employ daytime observations from CrIS onboard  
110 Suomi-NPP and focus on the western U.S. ( $30\text{-}50^\circ\text{N}$ ,  $99\text{-}127^\circ\text{W}$ ) during the summers of 2018  
111 (July 24 - September 13) and 2019 (July 22 - September 6); the specific timeframes match those  
112 for the airborne studies described in the next section.

113

114 The CrIS measurements of methanol, ethene, ethyne, and HCN employ the Retrieval of Organics  
115 from CrIS Radiances (ROCRv2) methodology described by Wells et al. (2025). Detection is  
116 based on the hyperspectral range index (HRI), a dimensionless quantity that quantifies the  
117 spectral signal for a given atmospheric species (Walker et al., 2011). HRIs for VOCs targeted  
118 here are derived over spectral ranges that encompass the  $\nu_8$  methanol absorption feature (C-O  
119 stretch;  $1020\text{-}1040\text{ cm}^{-1}$ ), the  $\nu_7$  ethene feature ( $=\text{CH}_2$  wag;  $940\text{-}960\text{ cm}^{-1}$ ), the  $\nu_5$  ethyne feature  
120 (C-H bend,  $720\text{-}740\text{ cm}^{-1}$ ), and the  $\nu_2$  HCN feature (H-C-N bend,  $700\text{-}720\text{ cm}^{-1}$ ). VOC column  
121 abundances are then quantified using an artificial neural network (ANN) retrieval that is trained



122 using synthetic HRIs generated by the Line-By-Line Radiative Transfer Model

123 (LBLRTMv12.13; Clough et al., 2005; Wells et al., 2025).

124

125 Relevant predictors for the trained ANN include, along with the CrIS-measured HRIs, variables

126 describing the atmospheric state (e.g., H<sub>2</sub>O column, surface pressure, skin and atmospheric

127 temperatures). Since thermal IR sensitivity depends on the thermal contrast between the Earth's

128 surface and the absorber, ROCRv2 includes an additional predictor ( $P_{90}$ ) that quantifies the

129 pressure below which 90% of the VOC column resides. The parameter is of particular

130 importance for smoke analyses given the potential for plumes to be lofted above the planetary

131 boundary layer and for subsequent altitude changes during transport.  $P_{90}$  is not known *a priori*

132 and is normally prescribed using independent information (e.g., from a model or from

133 independent observations); here, we use complementary pairs of observed VOCs to constrain

134 this vertical dependence.

135

136 Analyses in this paper primarily employ single-footprint HRIs and explore the vertical sensitivity

137 of these signals using the  $P_{90}$  metric. We additionally use CrIS-measured carbon monoxide (CO;

138 Worden et al., 2023; Kulawik and Payne 2025) as a fire tracer with an atmospheric lifetime far

139 exceeding that of individual smoke plumes. This CrIS CO dataset was generated using the

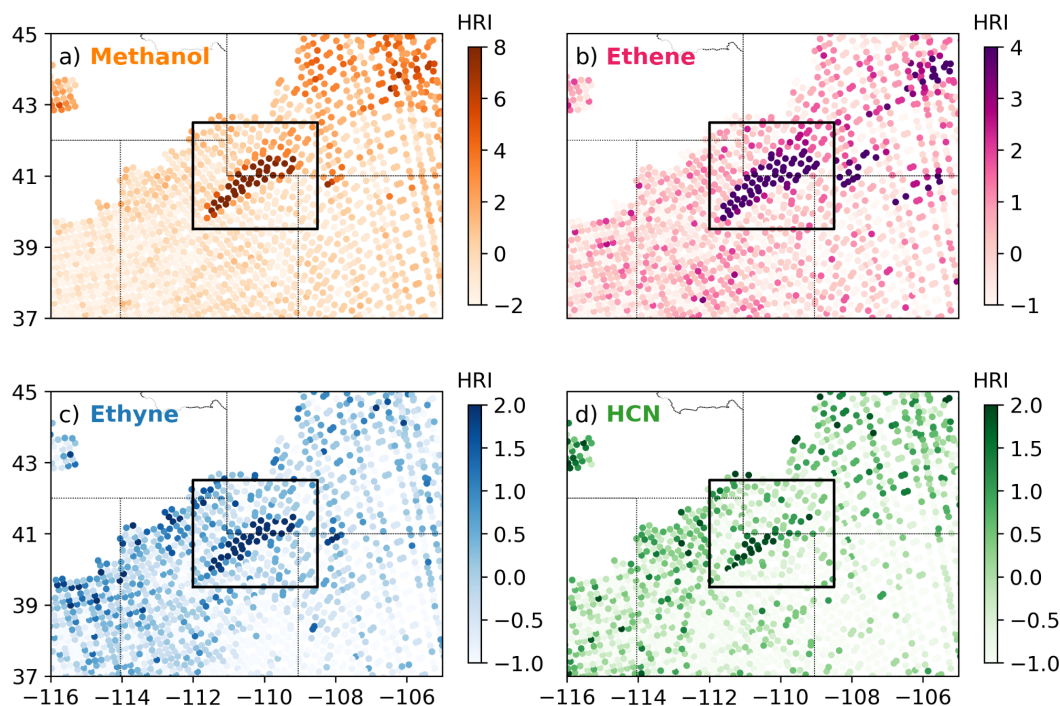
140 Tropospheric Ozone and Precursors for Earth System Sounding (TROPESS) processing system.

141 Figure 1 illustrates the HRIs for methanol, ethene, ethyne, and HCN observed by CrIS in the

142 Pole Creek fire plume on September 13, 2018 (Juncosa Calahorrano et al., 2021).

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145

146 **Figure 1.** CrIS spectral detection of a) methanol, b) ethene, c) ethyne, and d) HCN in an  
 147 individual fire plume. The hyperspectral range index (HRI) for each species is shown over a  
 148 portion of the western U.S. on September 13, 2018, with the black box delineating the Pole  
 149 Creek fire plume in Utah and Wyoming. Data gaps are due to clouds, which have been filtered  
 150 out following Wells et al. (2020).

151

## 152 **2.2 Airborne VOC observations in wildfire smoke plumes.**

153 The 2018 Western Wildfire Experiment for Cloud Chemistry, Aerosol Absorption and Nitrogen  
 154 (WE-CAN) (Lindaas et al., 2020; Juncosa Calahorrano et al., 2020; Permar et al., 2021) and the  
 155 2019 Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ)  
 156 (Warneke et al., 2022) airborne campaigns characterized the chemical composition and evolution  
 157 of biomass burning emissions over the western and southeastern U.S. WE-CAN (July 24 -  
 158 September 13, 2018) employed the NSF C-130 and included flights out of Boise, ID (16 flights)  
 159 and Broomfield, CO (3 flights). FIREX-AQ employed the NASA DC-8 and consisted of two  
 160 phases: the first (13 flights from July 22 - August 15, 2019) targeted western U.S. wildfires out  
 161 of Boise, ID, and the second (7 flights from August 21 - 31, 2019) targeted smaller fires in the



162 southeastern U.S. out of Salida, KS. Analyses here utilize data from WE-CAN and from the  
163 western portion of FIREX-AQ.  
164  
165 WE-CAN and FIREX-AQ featured VOC measurements using Proton-Transfer-Reaction Time-  
166 of-Flight Mass Spectrometers (PTR-ToF-MS) and Whole Air Samplers (WAS). The PTR-ToF-  
167 MS instruments enabled in-situ detection of VOCs with proton affinity higher than water and  
168 were operated at 1 or higher Hz sampling frequency. The WAS systems collected canister  
169 samples for subsequent off-line analysis. The NCAR Advanced-WAS (AWAS, Andrews et al.,  
170 2016) was deployed during WE-CAN (< 30 s sampling time; up to 46 canisters per flight), while  
171 the NOAA-integrated WAS (iWAS) was deployed during FIREX-AQ (< 10 s sampling time; up  
172 to 72 canisters per flight). Airborne analyses here employ PTR-ToF-MS methanol observations  
173 and AWAS/iWAS ethene observations, with all data averaged over the WAS sampling times.  
174 Uncertainties are reported at  $\pm 15\%$  (total; WE-CAN) and  $\pm 50\%$  (total; FIREX-AQ) for  
175 methanol, and at  $5\%/<8\%$  (accuracy/precision; WE-CAN) and  $2\%/5\%$  (accuracy/precision;  
176 FIREX-AQ) for ethene. More information about these instrument systems and their  
177 configuration during WE-CAN/FIREX-AQ can be found in Permar et al. (2021) and Warneke et  
178 al. (2022).

179

### 180 **2.3 Model description.**

181 We use the GEOS-Chem chemical transport model version 14.3.0 (The International GEOS-  
182 Chem User Community, 2021) to interpret the space-based VOC observations. The simulations  
183 use GEOS-FP assimilated meteorological fields from the NASA Global Modeling and  
184 Assimilation Office (GMAO), and are performed on a  $0.25^\circ \times 0.325^\circ$  grid nested over the  
185 western U.S. ( $33^\circ$ - $52^\circ$  N,  $99^\circ$  -  $129^\circ$ ). The nested runs extend from July 1 - September 30 of 2018  
186 and 2019, with initial and boundary conditions obtained from global simulations performed  
187 respectively at  $4^\circ \times 5^\circ$  and  $2^\circ \times 2.5^\circ$ . The model employs a hybrid-sigma vertical grid with 47  
188 layers extending to 0.01 hPa (22 in the lowest 5 km), and uses a 5-minute transport time step and  
189 a 10-minute chemical time step (10 and 20 minutes for the global runs). Model output is  
190 averaged from 12:00 - 15:00 LT for comparison with CrIS.

191



192 GEOS-Chem includes detailed HO<sub>x</sub>-NO<sub>x</sub>-VOC-ozone-BrO<sub>x</sub> chemistry coupled with aerosols.  
193 HCN is not a standard GEOS-Chem tracer and we include it here following Li et al. (2003;  
194 2009). Emissions in GEOS-Chem are computed using the HEMCO module, which includes  
195 biogenic VOC emissions from MEGANv2.1 (Guenther et al., 2012; Hu et al., 2015) and  
196 anthropogenic emissions from the Community Emissions Data System (CEDSV2). Jin et al  
197 (2023) evaluated multiple biomass burning inventories against the WE-CAN/FIREX-AQ  
198 airborne datasets and obtained the best model/measurement agreement when using the Global  
199 Fire Assimilation System (GFAS; CAMS, 2022); we therefore use GFAS here in the  
200 implementation described by Jin et al (2023). GFAS provides daily-averaged emissions for 40  
201 pyrogenic species by converting Fire Radiative Power (FRP) observations from the Moderate  
202 Resolution Imaging Spectroradiometer (MODIS; Justice et al., 2002; Giglio et al., 2003) into  
203 estimates of dry matter consumption; fluxes are then derived using literature-based emission  
204 factors (CAMS, 2022; Keiser et al., 2012). GFAS also includes a process-based parameterization  
205 of plume injection heights based on the heat and convective energy released from each fire  
206 (Latham et al., 1994). The mean altitude of maximum injection (“mami”) and the plume top are  
207 derived using a plume-rise model that is based on MODIS FRP and atmospheric profiles of  
208 temperature, wind, and atmospheric humidity from the European Center for Medium-Range  
209 Weather Forecasts (ECMWF; Freitas 2007, 2010; Paugam et al., 2015; Remy et al., 2017).  
210 Within GEOS-Chem, GFAS distributes fire emissions uniformly from the surface to the “mami”  
211 altitude while accounting for model resolution as described by Jin et al. (2023).

212

#### 213 **2.4 Smoke selection criteria.**

214 We use the CrIS CO and ethene measurements described earlier to define categories of smoke  
215 influence over the western U.S. Selection proceeds in two steps. We first impose a CO threshold  
216 (>150 ppb at 510 and 680 hPa; Fischer et al., 2018; Juncosa Calahorrano et al., 2021) to identify  
217 all scenes impacted by smoke during our study period. We then use the ethene observations to  
218 distinguish between dense and diluted smoke. Ethene is a good marker for this purpose because  
219 it is among the most abundant VOCs emitted from fires (Permar et al., 2021), and its short  
220 atmospheric lifetime (~6 h at [OH] = 5 × 10<sup>6</sup> molec/cm<sup>3</sup>; Atkinson et al., 2006) results in large  
221 near-source enhancements over a low background. That regional background differs between the  
222 two study years since 2018 was a far more active fire year (Jin et al., 2023), and we find that



223 different ethene HRI thresholds are therefore needed to resolve dense smoke scenes in the two  
224 cases (7 for 2018; 2 for 2019).

225

226 Finally, we aggregate the dense smoke scenes into coherent individual plumes for process  
227 analysis. Aggregation employs the density-based spatial clustering of applications with noise  
228 (DBSCAN) algorithm from the Python scikit-learn library (Ester et al., 1996; Schubert et al.,  
229 2017). DBSCAN groups data points with a dense population of close neighbors and excludes as  
230 outliers those that lie too far from a cluster. The two arguments needed for the algorithm are the  
231 radius distance and the minimum number of samples, which are set here to 60 km and 5,  
232 respectively (Figure S1). We next use fire detections from the Visible Infrared Imaging  
233 Radiometer Suite (VIIRS) with 375 m nominal resolution onboard Suomi-NPP to manually  
234 select coherent, intact plumes directly associated with a specific fire; we exclude transported  
235 plumes whose source location could not be readily identified (Schroeder et al., 2014a). The  
236 above procedure yields a total of 134 dense smoke plumes (116 in summer 2018; 18 in summer  
237 2019; Figure S2).

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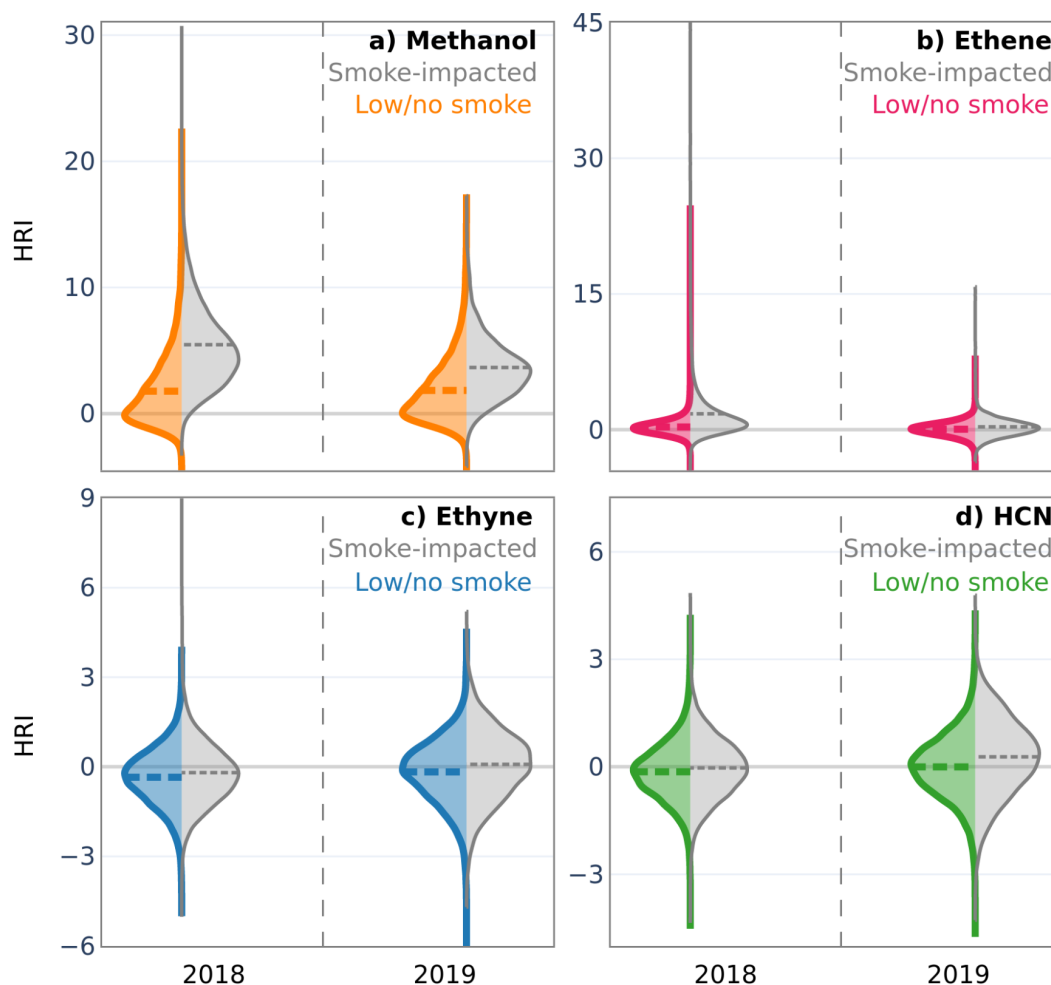


254 **Results and discussion.**

255 **Section 3.1. Methanol, ethene, ethyne, and HCN spectral enhancements in western U.S**

256 **smoke plumes.**

257



258

259 **Figure 2.** Distribution of VOC spectral signals in low/no-smoke and smoke-impacted scenes  
260 over the western U.S. as seen by CrIS during summers 2018 and 2019. Data reflect aggregated  
261 statistics at  $0.5^\circ \times 0.5^\circ$  horizontal resolution in low/no-smoke (color;  $N_{2018} = 38,214$ ;  $N_{2019} =$   
262  $27,848$ ) and smoke-impacted (grey;  $N_{2018} = 29,782$ ;  $N_{2019} = 3,694$ ) scenes for a) methanol, b)  
263 ethene, c) ethyne, and d) HCN. Violin plots show the spread and the probability density of the  
264 data based on kernel density estimators. The dashed line shows the mean of the distribution in



265 each case. Smoke categories are defined based on CO as described in the text. Figure S3 shows  
266 violin plots for the single-footprint retrievals.

267

268 Figure 2 summarizes the distribution of CrIS-observed spectral signals (HRIs) for methanol,  
269 ethene, ethyne, and HCN inside versus outside of smoke-impacted areas of the western U.S. By  
270 construction, the HRI has a mean of zero and a standard deviation of one under background  
271 conditions (Walker et al., 2011) so that some negative values are seen in all cases. The highest  
272 spectral signal in smoke-free scenes is observed for methanol, reflecting its additional biogenic  
273 sources and longer lifetime, and its status as one of the most abundant VOCs in the atmosphere.  
274 All four VOCs reveal a clear impact from fires, with peak  $0.5^\circ \times 0.5^\circ$  HRI values in smoke  
275 ranging from 4.1 for HCN to 44 for ethene (peak single-footprint values range from 7.5 for HCN  
276 to 125 for ethene; Tables S1 and S2). For comparison, two past studies have used space-based  
277 data to quantify VOC HRIs in smoke plumes. De Longeville et al. (2021) report single-footprint  
278 values for the Infrared Atmospheric Sounder Interferometer (IASI) downwind of the 2020  
279 Australian bushfires, with ethene HRIs exceeding 10 on four consecutive days. Wells et al.  
280 (2025) observed large VOC enhancements from the same Australian fires, with gridded CrIS  
281 HRI values (at  $0.5^\circ \times 0.625^\circ$ )  $>8$  for methanol and ethyne and  $>4$  for ethene and HCN. The  
282 specific HRI values vary with the spatial averaging approach and from instrument to instrument,  
283 as the signal depends strongly on the noise characteristics of the sensor (Hu et al., 2025).

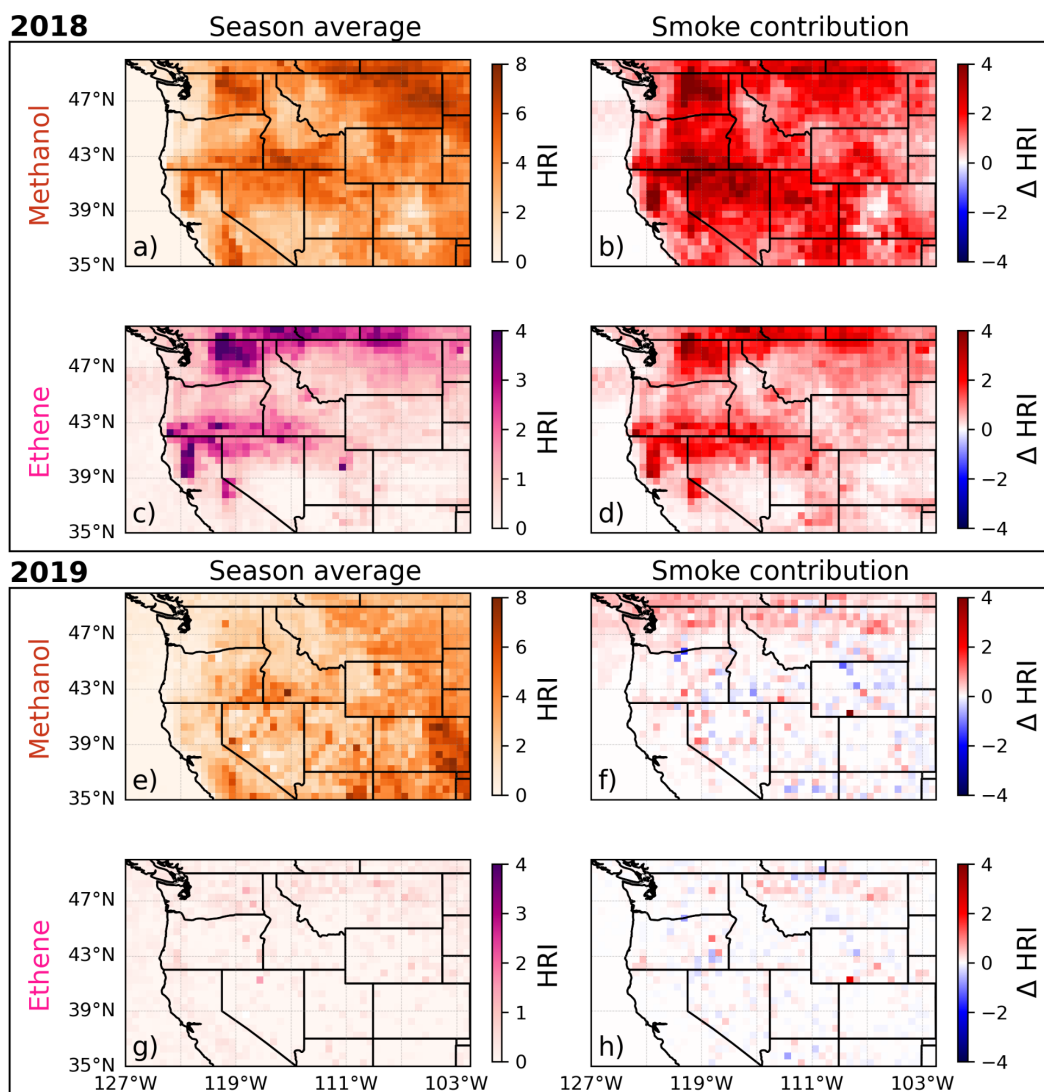
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285 The comparisons in Figure 2 also reveal larger peak smoke enhancements in 2018 than in 2019  
286 for all four VOCs. Peak in-smoke HRIs were 2-8 $\times$  higher (Table S1) in the more active 2018 fire  
287 season, which saw 6.7 million acres burnt across the western U.S. (Northwest, Northern Rockies,  
288 Northern California, Southern California, Great Basin, Southwest, and Rocky Mountain regions)  
289 versus just 1.6 million in 2019 (NIFC, 2018, 2019), resulting in a factor of 8.5 difference in the  
290 estimated total fire VOC emissions between the 2 years (Jin et al., 2023). This year-to-year  
291 difference is also reflected in the larger number of reported large fires for the region (560 in 2018  
292 vs. 395 in 2019). Large fires are defined as those that have burnt a minimum of 100 acres in  
293 timber and 300 in grass or brush type fuels (NIFC, 2018, 2019).

294



295 The peak spectral enhancements for methanol and ethene in smoke are much larger than those  
296 for ethyne and HCN (Figure 2). This order matches the speciation of fire emissions: methanol  
297 and ethene are among the top 10 pyrogenic VOCs, with emission factors for western U.S.  
298 ecosystems approximately five times those of ethyne and HCN (Burling et al., 2010, 2011;  
299 Gilman et al., 2015; Permar et al., 2021; Gkatzelis et al., 2024). However, spectroscopic effects  
300 also contribute to the observed HRI patterns. In particular, while the peak ethyne and HCN  
301 enhancements show clear impacts from fires, their mean in-smoke HRIs in 2018 are only  
302 modestly higher than the corresponding background values—in contrast to the situation for  
303 methanol and ethene (Table S1). In effect, some fire scenes exhibit strong spectral enhancements  
304 for all four VOCs, while others show strong enhancements for methanol and ethene but not for  
305 ethyne or HCN. The discrepancy arises because detection sensitivity varies with altitude to  
306 differing degrees among the four VOCs: this vertical dependence is particularly strong for ethyne  
307 and HCN because their absorption features lie near the  $720\text{ cm}^{-1}$   $\text{CO}_2$  band. In the following  
308 section, we explore implications of these species-dependent vertical sensitivities for fire VOC  
309 analyses.  
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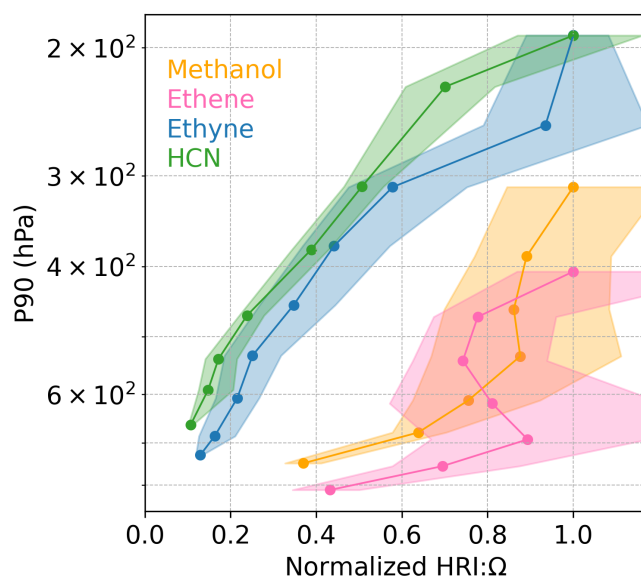
312 **Figure 3.** Spatial distribution of methanol and ethene spectral signals (HRIs) as measured by  
 313 CrIS over the western U.S. during the summers of 2018 and 2019. The left column shows the  
 314 seasonal-mean signals. The right column shows the contribution of smoke ( $\Delta$ HRI) to the  
 315 seasonal mean, quantified as the difference between the overall mean and the mean of all low/no  
 316 smoke observations (Figures S4 and S5). Data are plotted on a  $0.5^\circ \times 0.5^\circ$  grid and span the WE-  
 317 CAN (July 24 - September 13, 2018) and FIREX-AQ (July 26 - September 6, 2019) campaign  
 318 timeframes.



319 Figure 3 maps the seasonally-averaged HRIs for methanol and ethene over the western U.S.  
 320 during 2018/2019 campaign periods, and quantifies the contribution of smoke to that mean  
 321 signal. Larger smoke enhancements are seen in 2018, showing that during active fire years  
 322 pyrogenic VOCs are systematically elevated on a regional scale. These enhancements are  
 323 strongest near and downwind of persistent fires (e.g., the Carr and Mendocino Fires in  
 324 California) and are concentrated in the freshest portions of the plumes for ethene but more  
 325 dispersed for methanol—reflecting its longer lifetime ( $\sim 2.6$  d vs.  $\sim 6$  h at  $\text{OH} = 5 \times 10^6$  molec/cm<sup>3</sup>,  
 326 Atkinson et al., 2006). The California Central Valley exhibits large methanol signals in both  
 327 years with minimal smoke contribution, revealing the presence of important non-pyrogenic  
 328 sources such as dairy operations (e.g., Liu et al., 2022; Pfannerstill et al., 2023). The averaged  
 329 spatial HRI and  $\Delta$ HRI distributions for ethyne and HCN (Figure S4 and S5) are noisier than  
 330 those of methanol and ethene. While ethyne and HCN enhancements are clearly seen for  
 331 individual plumes (e.g., Figure 1), the impact of smoke on their seasonal-mean signals is less  
 332 evident due to their lower emissions and weaker strength-of-signal.

333

334 **3.2 Differences in thermal IR vertical sensitivity by compound**



335

336 **Figure 4.** CrIS measurement sensitivity (expressed as the spectral signal to column ratio, HRI:Ω)

337 as a function of the VOC's vertical location in the atmosphere. Data shown reflect model output



338 from GEOS-Chem/LBLRTM for smoky conditions in the western U.S. Results are shown for  
339 methanol, ethene, ethyne, and HCN as a function of  $P_{90}$ , the pressure below which 90% of the  
340 VOC column resides. Values are normalized to the peak sensitivity in each case, with lines and  
341 shading indicating the median and interquartile range for a given  $P_{90}$  bin.

342

343 Under typical conditions, the sensitivity of thermal IR measurements to a given VOC (i.e., the  
344 HRI:column ratio) increases with absorber altitude as the thermal contrast grows. Figure 4  
345 quantifies this effect for our target VOCs based on synthetic data from LBLRTM. The plotted  
346 data are filtered to be representative of smoke-impacted areas in the western U.S. Specifically,  
347 we include only overland scenes where the water vapor column falls within the range of CrIS-  
348 observed values ( $1.3 - 9.3 \times 10^{22}$  molecules  $\text{cm}^{-2}$ ) over our analysis domain. We also impose a  
349 minimum HRI threshold that corresponds to the mean observed in-smoke values for methanol  
350 (5.0) and ethene (1.2), and that is prescribed at 2.0 for ethyne and HCN given the low mean HRIs  
351 for these species. Finally, we require that the VOC-specific  $P_{90}$  values are bracketed (in 30 hPa  
352 bins) by the range encountered in western U.S. fire plumes based on GEOS-Chem output for  
353 2018 and 2019.

354

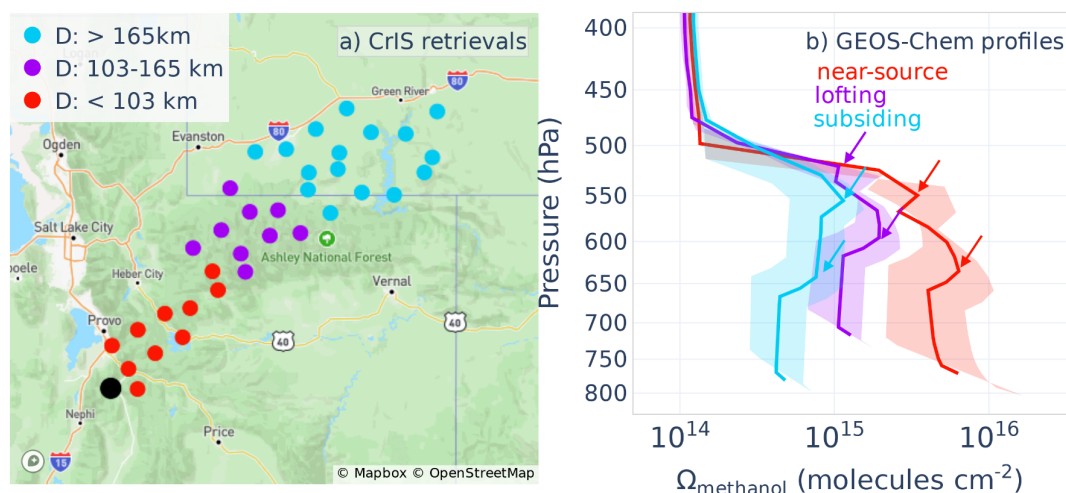
355 We see from Figure 4 that the vertical sensitivity dependence varies between compounds and is  
356 steeper for ethyne and HCN than it is for methanol and ethene, reflecting the  $\text{CO}_2$  effect  
357 discussed earlier. It is also evident from Figure 4 that the strength of this dependence is  
358 quantitatively similar between i) methanol and ethene, and ii) ethyne and HCN. This pairwise  
359 similarity raises the possibility of leveraging specific VOC combinations to support fire analyses  
360 in a way that addresses key uncertainties related to plume height. Specifically, we hypothesize  
361 that VOC pairs with similar vertical sensitivity will exhibit spectral correlations that are  
362 conserved downwind regardless of the plume's vertical location. As the plume rises or subsides,  
363 the VOC:VOC spectral ratio for such pairs should not change significantly because the  
364 sensitivities of the two species are varying similarly. Changes to this ratio during downwind  
365 transport will then only occur in response to processes that alter the plume's chemical  
366 composition. On the other hand, the spectral correlation between pairs of VOCs with differing  
367 vertical sensitivities will change as the plume moves vertically. Such changes may provide



368 information to constrain plume altitude changes during transport. In the following section, we  
 369 test these ideas using case-study observations from the Pole Creek Fire plume.

370

371 **3.3. Impact of plume altitude on VOC:VOC spectral ratios: the Pole Creek Plume.**



372

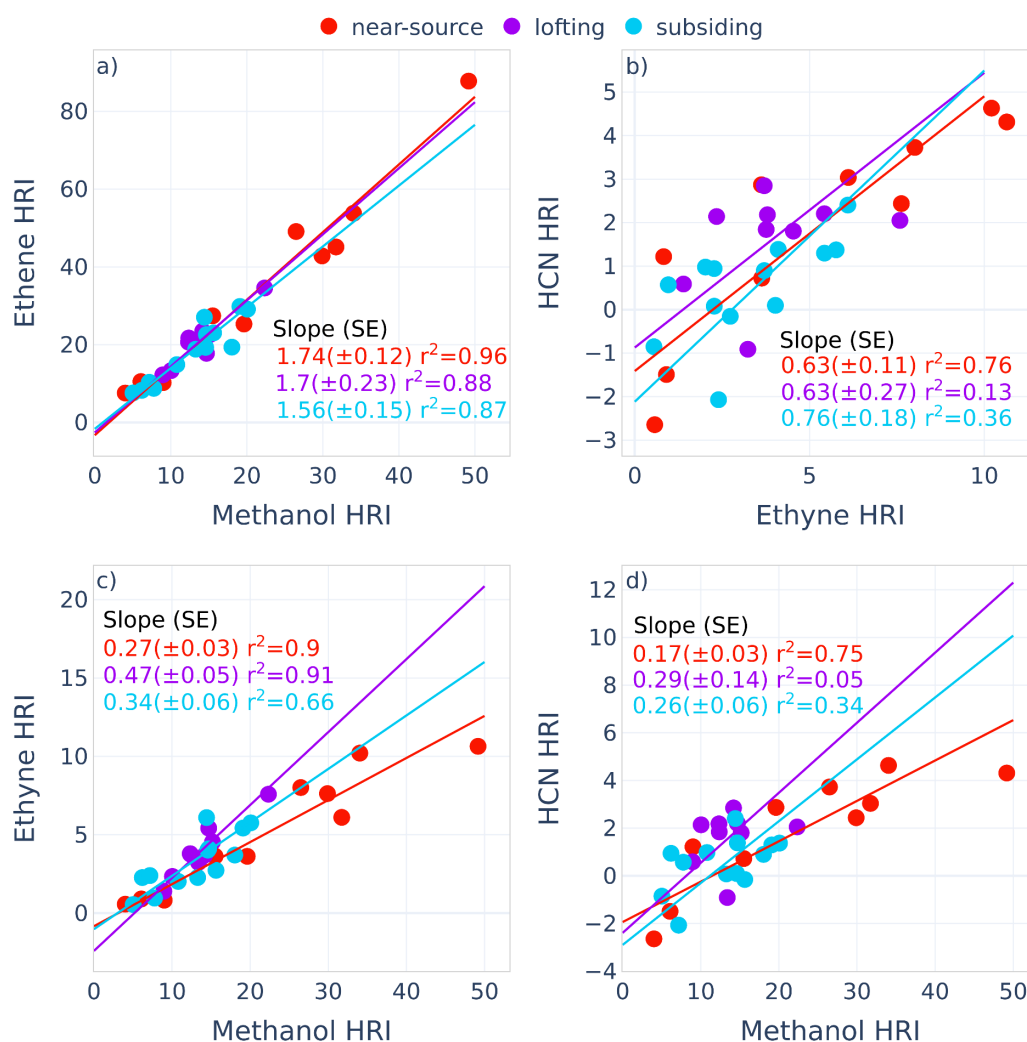
373 **Figure 5.** Plume transport and vertical displacement downwind of the Pole Creek Fire. a) CrIS  
 374 measurement locations within the Pole Creek Fire plume on September 13, 2018, grouped by  
 375 distance (D) from the fire centroid. The black dot shows the location of the VIIRS fire detection  
 376 at the source. b) Mean in-plume methanol profiles at the CrIS overpass time as simulated by  
 377 GEOS-Chem for the distance categories shown in panel a.

378

379 Biomass burning emissions rise vertically to an extent that depends on fire dynamics and on the  
 380 atmosphere's thermal stratification (Freitas et al., 2007, 2010, Paugam et al., 2015; Remy et al.,  
 381 2017). The resulting injection height is a highly uncertain model parameter and a focus of active  
 382 research (June et al., 2025; Singh et al., 2025). After initial injection, topography and convection  
 383 can lead to additional vertical displacement that further impacts the transport and fate of the  
 384 plume. The September 2018 Pole Creek Fire in central Utah provides a well-defined case study  
 385 for examining these processes (Juncosa Calahorrano et al., 2021). The lightning-initiated fire,  
 386 located in the Wasatch Range near Provo, gave rise to a distinct and coherent plume that  
 387 maintained its structure downwind with minimal influence from other regional sources. Clear  
 388 enhancements of methanol, ethene, ethyne, and HCN are evident in the CrIS observations of this



389 plume (Figure 1). Chemical transport model simulations and the regional topography (Figure 5)  
 390 indicate that after initial injection, the plume was lofted orographically as it passed through the  
 391 Uinta Mountains. The plume then subsided as it was transported past the Uinta Mountains and  
 392 into southwest Wyoming.  
 393



394  
 395 **Figure 6.** VOC spectral correlations as measured by CrIS in the Pole Creek Fire plume and as a  
 396 function of downwind distance. The top row plots pairs of VOCs with similar vertical  
 397 sensitivities; the slope of their spectral correlation does not statistically change with vertical



398 lofting. The bottom row shows pairs of VOCs with differing vertical sensitivities; here, the  
399 spectral signal for the VOC with more vertically-dependent sensitivity (plotted as the y-variable)  
400 is preferentially enhanced or suppressed as the plume is respectively displaced upward or  
401 downward. Distance categories reflect those shown in Figure 5. Slopes (with standard error) are  
402 calculated via Reduced Major Axis (RMA) regression.

403

404 Figure 6 plots the pairwise spectral correlations measured by CrIS for the four target VOCs  
405 within the Pole Creek Fire plume on September 13, 2018. The data are grouped by downwind  
406 distance and altitude as indicated in Figure 5. For methanol and ethene, with their similar vertical  
407 sensitivities, we observe a strong spectral correlation and a slope that is statistically independent  
408 of the plume's elevation. In fact, while the difference is not statistically significant, the lowest  
409 ethene:methanol slope is seen for the downwind population, which would be consistent with  
410 ethene oxidation over the ~4-5 h plume transport time (Juncosa Calahorrano et al., 2021). The  
411 same coherence holds for ethyne and HCN, the other VOC pair with similar vertical sensitivities,  
412 though with more scatter due to the weaker signal strength for these compounds. For such VOC  
413 pairs, the initial spectral slope at emission is thus unaffected by transport and will change only  
414 with the actual composition of the plume.

415

416 The situation is different for pairs of VOCs with divergent vertical sensitivities. Here, we expect  
417 the strength of the signal for the more vertically-sensitive compound (i.e., ethyne or HCN) to be  
418 preferentially enhanced as the plume rises and preferentially suppressed as the plume sinks. The  
419 VOC:VOC relationships plotted in Figure 6 (bottom row, with ethyne or HCN as the y-variable)  
420 are broadly consistent with this picture, with slopes that first increase as the plume is lifted over  
421 the Uinta Mountains, and then decrease as the plume subsides downwind. Chemical loss is not  
422 relevant in these cases given the corresponding oxidative lifetimes (from 2.3 d for ethyne to 74 d  
423 for HCN at  $\text{OH} = 5 \times 10^6 \text{ molec/cm}^3$ ; (Atkinson et al., 2006)).

424

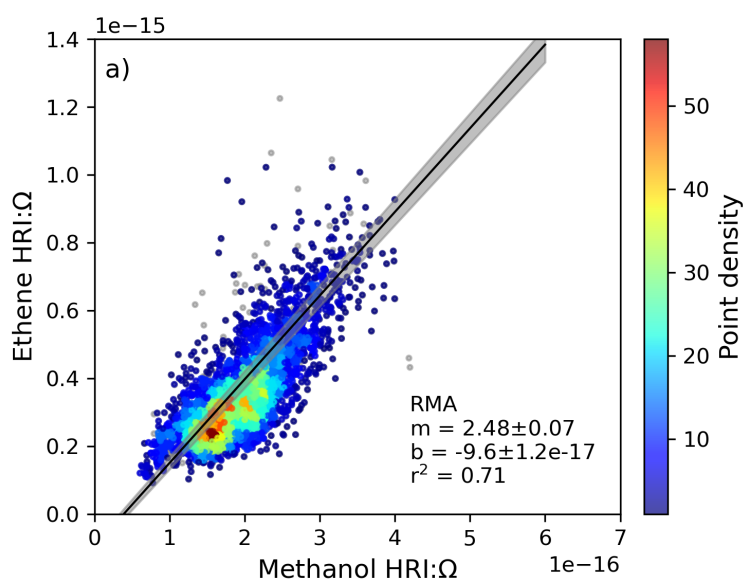
425 Species:species correlations are widely used in fire analyses to estimate emissions ratios with  
426 respect to a robust fire tracer (e.g., CO; Lindaas et al., 2020; Permar et al., 2021; Gkatzelis et al.,  
427 2024), to assess fire conditions and their evolution in time (Anderson et al., 2023), and to study  
428 downwind plume chemical evolution (Peng et al., 2020; Juncosa Calahorrano et al., 2020;



429 Lindaas et al., 2021; Jin et al., 2026). The findings above indicate that thermal IR spectral  
430 correlations between VOCs with different vertical sensitivities can help constrain plume altitude  
431 changes, while correlations between species with similar vertical sensitivities permit  
432 quantification of chemical ratios without requiring knowledge of the plume's vertical  
433 distribution. Below, we use CrIS observations to explore the second of these avenues with a  
434 focus on methanol and ethene.

435

### 436 3.4 Ethene:methanol column relationships in western U.S. wildfire plumes.



437

438 **Figure 7.** CrIS detection sensitivity for methanol versus ethene, plotted as the spectral signal to  
439 column ratio (HRI:Ω). Plotted values reflect synthetic data from the ROCRv2 training set,  
440 generated with LBLRTM based on randomized GEOS-Chem output. Data are filtered to reflect  
441 smoky conditions in the western U.S. and are colored by point density (N = 3090; 30 bins).  
442 RMA regression statistics with bootstrapped 95% confidence intervals are provided inset.

443

444 Quantifying VOC:VOC column enhancement ratios in smoke requires knowledge of their  
445 relative detection sensitivities—that is, their HRI:column (Ω) ratios. Figure 7 plots this quantity as  
446 computed by LBLRTM for methanol versus ethene, revealing the same close correlation ( $r^2 =$   
447 0.71) that was apparent in Figure 4. We can apply the corresponding slope as a conversion factor

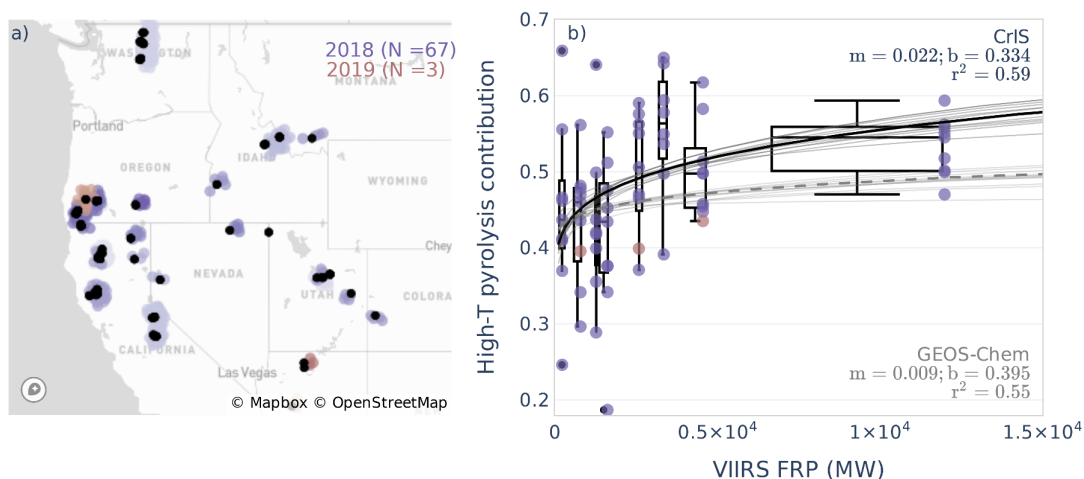


448 to directly translate CrIS-measured HRI:HRI enhancement ratios into column space for  
449 subsequent science analysis. For example, dividing the plume-wide Pole Creek methanol:ethene  
450 HRI slope of  $1.73 \pm 0.06$  by the sensitivity ratio from Figure 7 ( $2.48 \pm 0.04$ ) yields an  
451 ethene:methanol column enhancement ratio of  $0.70 \pm 0.03$ . The derivation requires no prior  
452 knowledge of the plume's vertical location and is thus not sensitive to uncertainties in plume  
453 injection height or to any subsequent altitude changes during transport. It also avoids relying on  
454 the neural-network retrieval used for global CrIS analyses (Wells et al., 2025), in which we find  
455 these extremely concentrated plume conditions are out-of-sample.

456

457 We next apply the above methodology to compute ethene:methanol column enhancement ratios  
458 for the set of dense wildfire plumes detected by CrIS over the western U.S. during the summer of  
459 2018 and 2019. The analysis includes all plumes meeting the dense smoke criteria defined earlier  
460 along with the following requirements: i)  $> 4$  CrIS observations; ii) ethene:methanol HRI  
461 correlation of  $r^2 > 0.4$ ; iii) slope uncertainty  $< 80\%$ ; and iv)  $< 75$  km from the fire source. The  
462 last requirement constrains the plume transport time to  $< 2.1 (\pm 0.8)$  h at an average wind speed  
463 of  $10 (\pm 4)$   $\text{m s}^{-1}$ , and is imposed to limit the impact of ethene oxidation on the derived ratios. In  
464 some of the cases outflow from 2-3 proximate fires merged into a single plume. When such  
465 sources were more than 100 km apart we considered them to be independent and applied the  
466 selection conditions separately to each one (some observational overlap is possible in this case).  
467 Outflow from fires less than 100 km apart was considered as a single plume, with the minimum  
468 possible downwind distance used in applying the near-source selection criterion. In this way we  
469 obtain a total of 67 plumes spanning 39 days during summer 2018, and 3 plumes spanning 3 days  
470 during summer 2019 (Figure 8a).

471



472

473 **Figure 8.** a) Fresh fire plumes detected by CrIS over the western U.S. during the summers of  
 474 2018 (67; purple) and 2019 (3; copper). Black dots show the averaged locations across all VIIRS  
 475 detections of each fire. b) Contribution of high-temperature pyrolysis for each of these fire  
 476 plumes (estimated from the CrIS ethene:methanol ratios as described in-text) as a function of  
 477 VIIRS fire radiative power (FRP). Boxplots encompass the interquartile range (IQR) for each  
 478 data bin; whiskers span data points within  $1.5 \times$  IQR of the first and third quartiles. Boxplots are  
 479 positioned at the bin-median FRPs with widths indicating the corresponding standard deviations.  
 480 Lines of best fit with regression coefficients listed inset reflect a fourth-root power-law  
 481 relationship and are calculated using the median high-temperature contribution and FRP for each  
 482 bin. Thin grey lines show results from sensitivity analyses varying the plume selection criteria  
 483 and number of analysis bins.

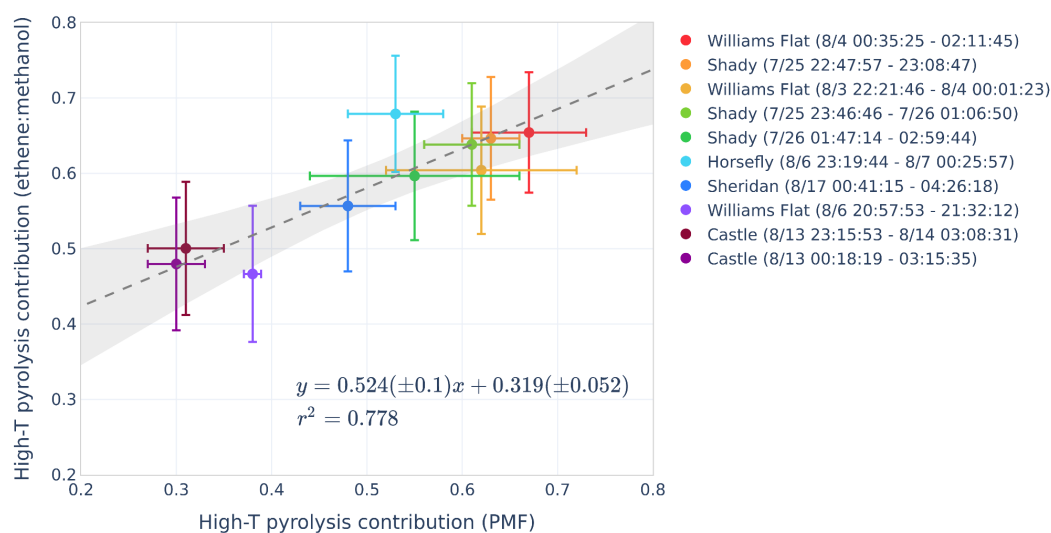
484

485 The CrIS-observed ethene:methanol column enhancement ratios for the 70 plumes have a mean  
 486 value of 0.61 (standard deviation:  $\pm 0.23$ ), which is very similar to the mean GFAS emission  
 487 ratio over this time and domain ( $0.63 \pm 0.08$ ; Figure S6). Both values are lower than the mean in-  
 488 situ emission ratios across the ensemble of plumes sampled during the airborne WE-CAN  
 489 ( $1.23 \pm 0.73$ ; Permar et al., 2021) and FIREX-AQ ( $1.25 \pm 0.41$ ; Gkatzelis et al., 2024) campaigns.  
 490 A model test shows that in-situ pyrogenic ethene:methanol ratios can be directly compared to the  
 491 corresponding total column enhancement ratios, and that both are expected to be similar to the  
 492 underlying emission ratio (Figure S7). The comparison above thus indicates that the airborne



493 sampling encountered plume conditions with higher ethene:methanol ratios than occur in the 70-  
 494 plume CrIS dataset. The airborne sampling generally took place 2-4 hours after the CrIS  
 495 overpass time, during the diel peak in fire activity and FRP (Wiggins et al., 2020); as will be  
 496 seen below, higher ethene:methanol ratios are expected at that time. Both the airborne data and  
 497 the CrIS observations also reveal a greater degree of ethene:methanol variability (standard  
 498 deviations of 0.23-0.73) than would be predicted solely from the GFAS emission ratios (0.08).  
 499  
 500 Fuel type is traditionally the only variable considered when parameterizing fire VOC speciation  
 501 in models and inventories (Andreae and Merlet, 2001; Akagi et al., 2011; Keiser et al., 2012;  
 502 Andreae 2019). Dominant fuel types in the western U.S. include boreal and temperate forests,  
 503 savannas, and cropland (Keiser et al., 2012), with reported ethene:methanol emission ratios of  
 504 0.53-0.70, 0.65 - 0.74, and 0.19 - 0.47, respectively (Andreae and Merlet, 2001; Akagi et al.,  
 505 2011; Andreae 2019). The GFAS predictions for our analysis domain fall within that overall  
 506 range, with fluxes mainly occurring at two discrete emission ratios of 0.61 and 0.66 (Figure S6).  
 507 The larger spread seen in the spaceborne and in-situ observations points to other factors  
 508 influencing pyrogenic VOC speciation within and between fuel types, and we investigate one  
 509 such factor below.

510



511



512 **Figure 9.** Fire pyrolysis conditions as inferred from FIREX-AQ airborne observations. High-  
513 temperature pyrolysis fractions derived from the in-plume ethene:methanol ratios (plotted as the  
514 y-variable) track the positive matrix factorization (PMF)-based estimates from Sekimoto et al.  
515 (2023) (plotted as the x-variable).

516

517 Recent analyses suggest that differences in pyrogenic VOC speciation arise mainly from distinct  
518 pyrolysis conditions. In particular, Sekimoto et al. (2018, 2023) found via positive matrix  
519 factorization (PMF) that 70-85% of the VOC variability across western U.S. fuels could be  
520 explained by two factors representing high-temperature and low-temperature pyrolysis processes.  
521 In effect, each factor is an averaged emission profile (ppb VOC/ppb total measured VOCs) that  
522 provides a fingerprint for high-T or low-T conditions across hundreds of VOCs. They further  
523 showed that these high-T versus low-T contributions can be estimated from the observed ratios  
524 of individual VOCs that are emitted under contrasting pyrolysis conditions (Eq. S1, S2). The  
525 example ratio they examined was that of ethyne (mainly a high-T product) to furan (low-T).  
526 However, neither compound is routinely observed outside of dedicated in-situ campaigns, and  
527 furan is very short-lived ( $\sim 1$  h at  $[\text{OH}] = 5 \times 10^6$  molec/cm<sup>3</sup>; (IUPAC, 2024)), hindering broader  
528 application.

529

530 Ethene and methanol also have contrasting pyrolysis profiles, with ethene predominantly (72%)  
531 emitted under high-T conditions and methanol predominantly (62%) at low-T (Sekimoto et al.,  
532 2018). This complementarity, and the availability of space-based observations for both species,  
533 motivates us to explore the utility of the ethene:methanol pair for inferring high-T vs. low-T  
534 VOC emission contributions. Figure 9 tests this idea using airborne data from FIREX-AQ,  
535 comparing the high-T contribution as derived from in-plume ethene:methanol ratios (Eq. S2) to  
536 the Sekimoto et al. (2023) PMF-based results for their 10-plume dataset. The two estimates track  
537 each other closely ( $r^2 = 0.78$ ), with a non-zero intercept that may reflect the limitations of a 2-  
538 species solution versus one based on hundreds of VOCs. Nevertheless, the comparison supports  
539 the idea that the ethene:methanol ratio is well-suited for characterizing the dominant thermal  
540 process occurring in fires—provided that observations are restricted to the near-field given  
541 ethene's  $\sim 6$  h lifetime. We therefore extend the analysis above to the CrIS-observed



542 ethene:methanol column enhancement ratios, which offer the opportunity for fire  
543 characterization that goes beyond the coverage of in-situ observations.

544

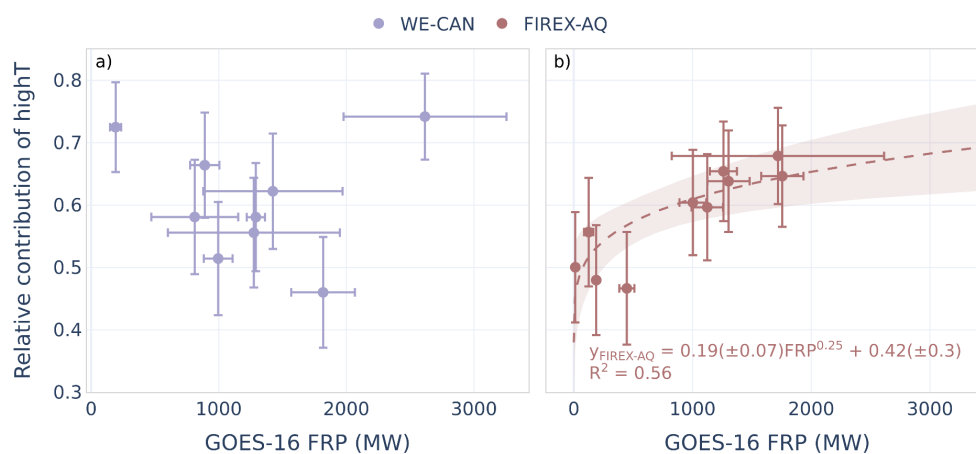
545 Figure 8b shows the high-T pyrolysis contributions derived from CrIS ethene:methanol column  
546 enhancement ratios for the population of fresh plumes (N=70) observed during the summers of  
547 2018 and 2019 over the western U.S. Although these plumes were all measured at similar times  
548 of day and originate from similar fuel types, they exhibit a wide range of ethene:methanol ratios  
549 (0.15-1.28; Figure S8) and therefore of inferred high-T pyrolysis contributions (0.19-0.67). Data  
550 in Figure 8b are plotted as a function of VIIRS FRP, which as a measure of the energy released  
551 by a fire is proportional to the rate of fuel consumption and to the fourth power of absolute  
552 temperature (Wooster et al., 2003). Higher FRP values indicate more intense flaming and  
553 greater heat release, creating favorable conditions for high-T pyrolysis and therefore for a greater  
554 proportion of high-T VOCs (Sekimoto et al., 2023). For each fire we compute the corresponding  
555 FRP as the sum of all values within that fire scar for the corresponding CrIS measurement time;  
556 plumes with multiple fire sources then use the mean FRP across sources. We account for any  
557 bow-tie effects (Wolfe et al., 2013) by removing one observation from FRP pairs separated by <  
558 375 m.

559

560 The high-T pyrolysis fractions inferred from the CrIS data increase with FRP at a rate of 0.022  
561 (0.014 - 0.026) MW<sup>-1</sup> with  $r^2 = 0.59$  (0.35 - 0.71). Values in parentheses reflect the range across  
562 a suite of sensitivity tests varying the number of analysis bins (5-9) and the plume selection  
563 criteria (ethene:methanol HRI correlation threshold  $r^2 = 0.4-0.6$ ; slope uncertainty 60-80%;  
564 maximum distance from source 50-125 km; fire separation distance 75-150 km). Results indicate  
565 that the observed relationship is statistically robust. Simulated results from GEOS-Chem  
566 (computed from the ethene:methanol column enhancement ratios in the same manner) show a  
567 similar correlation ( $r^2 = 0.55$  [0.19 - 0.87]; Figure 8b) but a weaker slope (0.009 [0.002 - 0.012]  
568 MW<sup>-1</sup>). While the magnitude of modeled emissions (and injection altitude) for any given fire is  
569 computed in GFAS as a function of FRP, the associated VOC speciation is based solely on fuel  
570 type without any explicit FRP dependence. The model correlation seen in Figures 8b and S8 thus  
571 arises solely from an association between fire FRP and vegetation fuel type. We propose that the  
572 model slope underestimate arises, at least in part, from the fact that higher FRP values also drive



573 enhanced high-T pyrolysis within individual vegetation categories, whereas GFAS (like other  
 574 fire emission inventories) uses static emission factors for the entire fire life cycle.  
 575



576  
 577 **Figure 10.** Fire pyrolysis conditions inferred from in-situ ethene:methanol observations as a  
 578 function of FRP as measured by GOES West (GOES-16) in wildfire plumes over the western  
 579 U.S. Plumes sampled during the a) 2018 WE-CAN campaign and b) 2019 FIREX-AQ campaign  
 580 are shown.

581  
 582 Sekimoto et al. (2023) found a similarly strong correlation between their PMF-derived high-T  
 583 pyrolysis contribution and FRP (from GOES-East and GOES-West) for their set of plumes  
 584 sampled in-situ during FIREX-AQ. We find that this correlation is maintained when instead  
 585 deriving the high-T contribution from in-situ ethene:methanol ratios (Figure 10b). However, the  
 586 same coherence is not apparent in the WE-CAN in-situ data from the prior year. In particular, an  
 587 analysis of the 9 WE-CAN plumes with < 4 h physical age featuring the most robust sampling  
 588 (Permar et al., 2021) reveals no correlation between FRP and either the in-situ ethene:methanol  
 589 ratios or the corresponding high-T pyrolysis fractions (Figure 10a). This WE-CAN comparison is  
 590 restricted to FRP values from GOES-West since GEOS-East was not operational during summer  
 591 2018, and differences should also be expected with respect to the CrIS VIIRS-based analysis  
 592 since FRP estimates differ between sensors (Li et al., 2018; 2020). Nevertheless, the WE-CAN  
 593 FRP results appear at odds with the CrIS and FIREX-AQ findings. Possible explanations include  
 594 the following: 1) the widespread regional smoke during the WE-CAN timeframe (Figure 3) may



595 affect the observed ethene:methanol ratios; 2) the more extensive smoke in 2018 could  
596 contribute to fire omission errors or to FRP underestimates (Schroeder et al., 2014b); 3) the  
597 sparser ethene observations during WE-CAN could prevent accurate diagnosis of the in-plume  
598 ethene:methanol ratios; or 4) the relationship between FRP, ethene:methanol, and pyrolysis  
599 conditions may be more variable than is apparent solely from the CrIS spaceborne and FIREX-  
600 AQ airborne data. More research is needed to clarify this issue and the broader utility of space-  
601 based ethene:methanol observations for fire process analysis.

602

#### 603 **4. Conclusions**

604 Despite extensive research on wildfires, important uncertainties still exist regarding the chemical  
605 speciation of emitted compounds—including VOCs, which drive formation of aerosols and ozone.  
606 Satellite observations in the thermal IR provide new avenues for investigating the chemical  
607 properties of smoke, as they are sensitive to a variety of fire VOCs even in aerosol-rich plumes.  
608 Here, we analyzed CrIS observations of four pyrogenic VOCs (methanol, ethene, ethyne, HCN)  
609 in wildfire smoke over the western U.S during the summers of 2018 and 2019, corresponding  
610 respectively to the timeframes of the WE-CAN and FIREX-AQ airborne campaigns. We  
611 explored the use of such data for fire process studies, with particular focus on the impacts of  
612 vertically-dependent detection sensitivity and on leveraging species:species ratios to minimize  
613 plume-height uncertainties and diagnose fire properties.

614

615 The CrIS VOC spectral signals are strongly enhanced in wildfire smoke plumes, and these  
616 enhancements are larger for the more abundantly-emitted VOCs (i.e., methanol and ethene) and  
617 during more active fire seasons (i.e., 2018). Enhancements for shorter-lived VOCs can be used to  
618 identify fresh smoke, while those for longer-lived species map wider-range patterns of plume  
619 transport. Fires were widespread in 2018, and smoke impacts dominated the seasonal-mean  
620 spectral signals for the analyzed VOCs across the western U.S. The same did not occur in 2019,  
621 when there were far fewer fires. We find that thermal IR observations of ethene are well-suited  
622 for fire plume identification in both scenarios. As a short-lived pyrogenic VOC with low  
623 background concentrations it provides a clearer fingerprint of fresh smoke than do longer-lived  
624 tracers (e.g. CO, Latham et al., 2013; Kloss et al., 2021; Arnold et al., 2025), while avoiding the



625 need for manual visual analysis (e.g. NOAA Hazard Mapping System, Shen et al., 2026; Lee &  
626 Jaffe, 2024).

627

628 We show that fire-emitted VOCs with similar vertical sensitivities exhibit tight spectral  
629 (HRI:HRI) correlations that remain unchanged even if a plume changes altitude during transport.  
630 Plume injection heights are among the most uncertain parameters for wildfire modeling (Paugam  
631 et al., 2016): for instance, the GFAS inventory features observationally-informed plume height  
632 estimates, but recent studies find that a 1.7-2× increase in these values is needed to match  
633 regional and global aerosol observations (Singh et al., 2025; June et al., 2025). Findings here  
634 provide a methodology for deriving VOC:VOC column enhancement ratios from space-based  
635 data without requiring any information on plume height.

636

637 Conversely, for VOC pairs with differing vertical sensitivities, the HRI:HRI correlation changes  
638 in a coherent way as a function of plume altitude. Such VOC pairs could potentially be used to  
639 constrain near-source injection heights or subsequent plume altitude changes. A more detailed  
640 investigation of these effects, and their sensitivity to other factors (e.g., to any vertically-varying  
641 interferences) is needed to explore the broader viability of such an approach.

642 We next quantified ethene:methanol emission ratios across 70 western US fire plumes detected  
643 by CrIS during 2018 and 2019. The mean molar emission ratio measured by CrIS (0.61) is  
644 similar to the bottom-up prediction from GFAS (0.63) for the same region and time periods, but  
645 the satellite observations reveal a greater degree of variability than is present in the inventory  
646 (standard deviations of 0.23 vs. 0.08). The satellite-based emission ratios are lower than those  
647 reported by the WE-CAN and FIREX-AQ airborne field campaigns (~1.24); this difference may  
648 reflect the 2-4 h later in-situ sampling time, which coincides with peak fire intensity and likely  
649 with greater ethene:methanol emission ratios.

650

651 Finally, we present a satellite-based proxy approach for assessing the thermal conditions  
652 affecting fire VOC emissions. Specifically, we show that the relative importance of high- versus  
653 low-temperature pyrolysis conditions in a fire can be inferred from the in-plume ethene:methanol  
654 ratios. Pyrolysis fractions derived in this way correlate well with in-situ PMF-based results that  
655 are informed by hundreds of different VOCs (Sekimoto et al., 2018, 2023); space-borne



656 ethene:methanol observations could therefore be broadly useful for constraining VOC speciation  
657 in wildfire plumes. We further demonstrate that high-T pyrolysis contributions derived from  
658 CrIS-observed ethene:methanol column enhancement ratios correlate with the fire radiative  
659 power (FRP) at the time of emission, as expected given the association between FRP and burn  
660 conditions that favor higher-T processes. Such a correlation also manifests in model predictions  
661 as a result of the association between FRP and fuel type; however, in that case the slope of the  
662 dependence is weaker and we suggest that this disparity reflects the fact that FRP also affects fire  
663 VOC speciation within individual ecosystem categories. A strong ethene:methanol vs. FRP  
664 correlation is apparent in airborne observations from the 2019 FIREX-AQ campaign, matching  
665 the CrIS findings, but the same is not true for the 2018 WE-CAN study which encountered  
666 widespread diluted smoke. The disparity underscores a need for additional analyses to  
667 understand drivers of VOC speciation under both fresh and aged smoke conditions.

668

669 Overall, these results demonstrate the utility of space-based VOC:VOC data for fire process  
670 analysis. A range of pyrogenic species with complementary chemical and detection properties  
671 are observable in the thermal IR. This opens opportunities for characterizing plume chemistry  
672 and transport, and for improving emission parameterizations in satellite-informed inventories  
673 (e.g., those using FRP). Findings here focus on the western U.S.; other sources of variability may  
674 play a role in different ecosystems, and additional research is needed to develop a more  
675 generalized understanding of fire VOC processes across different ecosystems and thermal  
676 regimes.

677

#### 678 **Data availability**

679 The ROCRv2 VOC data used in this work are available at <https://doi.org/10.13020/9r8x-pp66>  
680 (Wells et al., 2025). The single-footprint CrIS CO data used in this work are available at  
681 <https://zenodo.org/records/17065859> (2018; Kulawik and Payne, 2025) and  
682 <https://conservancy.umn.edu/items/c480f4b1-bb73-4612-81ac-388b468583dd> (2019; Juncosa  
683 Calahorrano et al., 2026). The GEOS-Chem model code can be obtained at Zenodo  
684 <https://doi.org/10.5281/zenodo.5711194>. WE-CAN data is archived at  
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690 Team, 2020).

691

#### 692 **Code availability**

693 <https://conservancy.umn.edu/items/c480f4b1-bb73-4612-81ac-388b468583dd> (Juncosa  
694 Calahorrano et al., 2026)

695

696 **Author contributions.** Conceptualization: DBM and KCW. Data curation: JFJC, KCW, CH,  
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701

#### 702 **Competing interests**

703 One of the co-authors is a member of the editorial board for Atmospheric Chemistry and Physics.  
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705

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