Supplementary Information

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

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Section S1. High-temperature pyrolysis contribution as derived from ethene:methanol

Following the Sekimoto et al. (2018) formulation for ethyne and furan, the ratio R of high-temperature to low-temperature pyrolysis contributions can be estimated as:

$$R = \frac{High-T}{Low-T} = \frac{ethene/A}{methanol/B} \quad \text{Eq. S1}$$

where A (0.0595 ppb ethene/ppb total VOC) and B (0.0946 ppb methanol/ppb total VOC) are the average emission factors under high-T and low-T conditions.

The fractional contribution F of high-T pyrolysis can then be derived as:

$$F = \frac{High-T}{High-T + Low-T} = \frac{R}{1+R} \quad \text{Eq. S2}$$

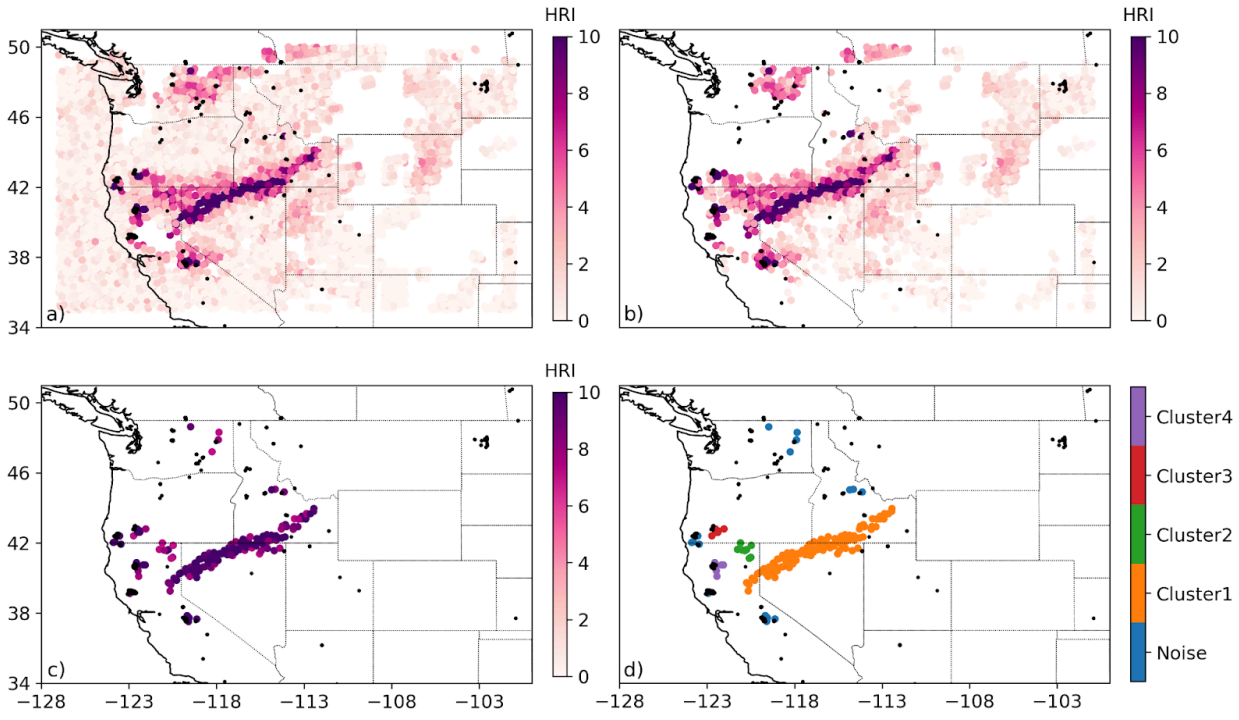


Figure S1. Illustration of smoke selection criteria for August 3, 2018. a) All CrIS ethene observations. b) Smoke-impacted scenes as defined by CrIS CO (>150 ppb at 510 and 680 hPa). c) Dense smoke plumes defined using an additional filter based on CrIS ethene (HRI > 7). d) Distinct plume clusters derived using DBSCAN. Black dots show VIIRS daytime fire detections for the same day.

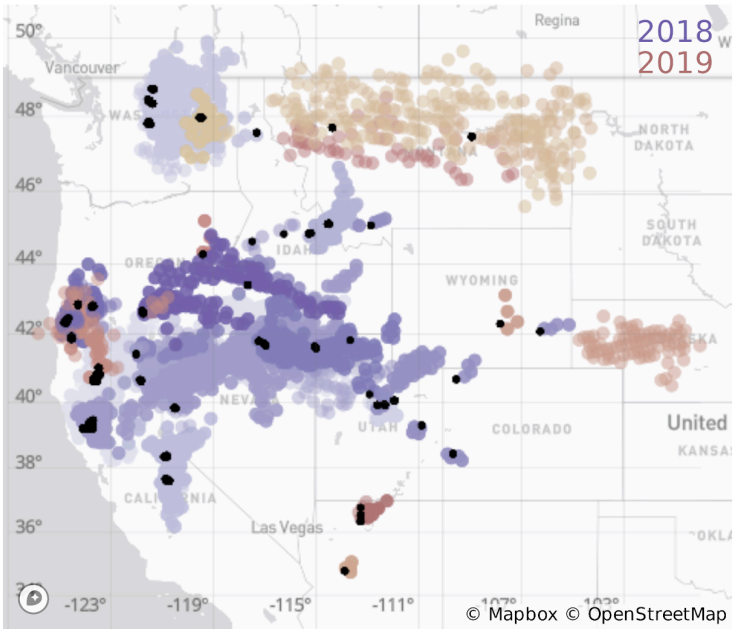


Figure S2. Dense plumes identified using CrIS observations of CO and ethene, grouped via the DBSCAN clustering algorithm. Purple and copper plumes indicate observations from 2018 and 2019, respectively. Black dots show the corresponding VIIRS fire detections.

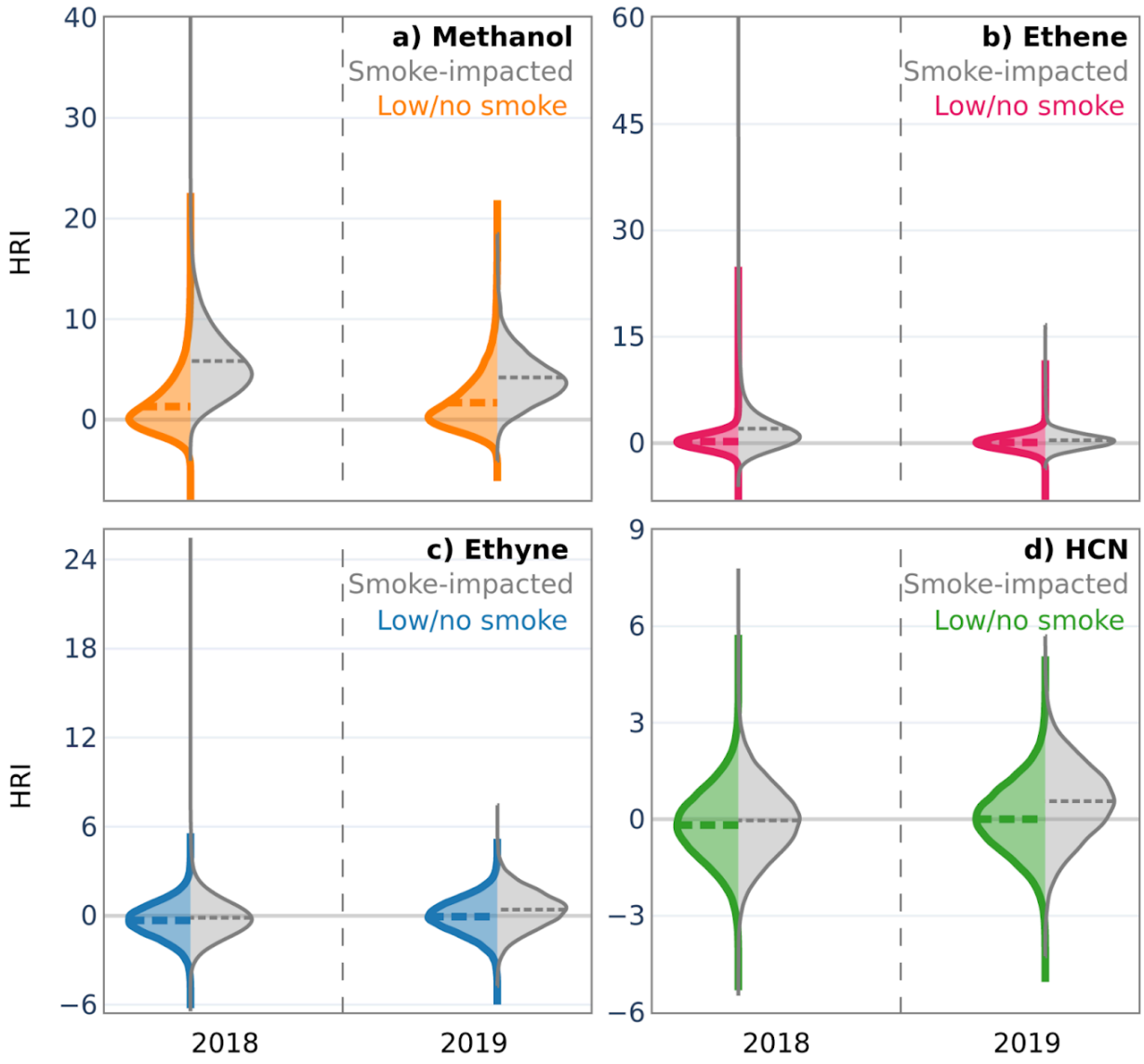


Figure S3. Distribution of VOC spectral signals in low/no-smoke and smoke-impacted scenes over the western U.S. as seen by CrIS during summers 2018 and 2019. Data reflect single-footprint statistics in low/no smoke (color; N_{2018} = 158,237; N_{2019} = 113,044) and smoke-impacted (grey; N_{2018} =108,477; N_{2019} = 7,424) scenes for a) methanol, b) ethene, c) ethyne, and d) HCN. Violin plots show the spread and the probability density of the data using kernel density estimators. The dashed line shows the mean of the distribution in each case. Smoke categories are defined based on CO as described in the text. Figure 2 shows violin plots for the same data aggregated to 0.5º × 0.5º horizontal resolution.

Table S1. Aggregated (0.5º × 0.5º) CrIS HRI statistics in low/no-smoke and smoke-impacted scenes during the summers of 2018 and 2019

	2018		2019	
	Low/no-smoke mean (±std error) [range]	Smoke-impacted mean (±std error) [range]	Low/no smoke mean (±std error) [range]	Smoke-impacted mean (±std error) [range]
Methanol	1.78 (±0.01) [-4.52, 21.97]	5.47 (±0.02) [-2.50, 29.89]	1.84 (±0.01) [-4.44, 16.76]	3.66 (±0.04) [-3.26, 16.36]
Ethene	0.29 (±0.01) [-4.29, 24.24]	1.75 (±0.02) [-3.61, 44.24]	0.05 (±0.01) [-6.30, 7.92]	0.32 (±0.02) [-3.14, 15.41]
Ethyne	-0.35 (±0.01) [-4.77, 3.79]	-0.20 (±0.01) [-4.75, 8.78]	-0.17 (±0.01) [-5.75, 4.35]	0.08 (±0.02) [-4.20, 4.70]
HCN	-0.14 (±0.01) [-4.33, 4.05]	-0.03 (±0.01) [-4.12, 4.62]	0.00 (±0.01) [-4.53, 4.15]	0.28 (±0.02) [-4.53, 4.15]

Table S2. Single-footprint CrIS HRI statistics in low/no-smoke and smoke-impacted scenes during the summers of 2018 and 2019

	2018		2019	
	Low/no smoke mean (±std error) [range]	Smoke-impacted mean (±std error) [range]	Low/no-smoke mean (±std error) [range]	Smoke-impacted mean (±std error) [range]
Methanol	1.29 (±0.01) [-7.70, 21.97]	5.83 (±0.01) [-2.91, 56.59]	1.68 (±0.01) [-5.58, 21.29]	4.19 (±0.03) [-3.26, 17.62]
Ethene	0.24 (±0.01) [-7.71, 24.24]	2.06 (±0.01) [-3.61, 124.39]	0.09 (±0.01) [-8.79, 11.28]	0.42 (±0.02) [-3.14, 16.28]
Ethyne	-0.31 (±0.01) [-6.01, 5.34]	-0.13 (±0.01) [-5.82, 24.84]	-0.05 (±0.01) [-5.75, 4.94]	0.42 (±0.02) [-4.26, 6.99]
HCN	-0.18 (±0.01) [-5.10, 5.53]	-0.04 (±0.01) [-5.23, 7.55]	0.00 (±0.01) [-4.84, 4.85]	0.56 (±0.01) [-4.84, 4.85]

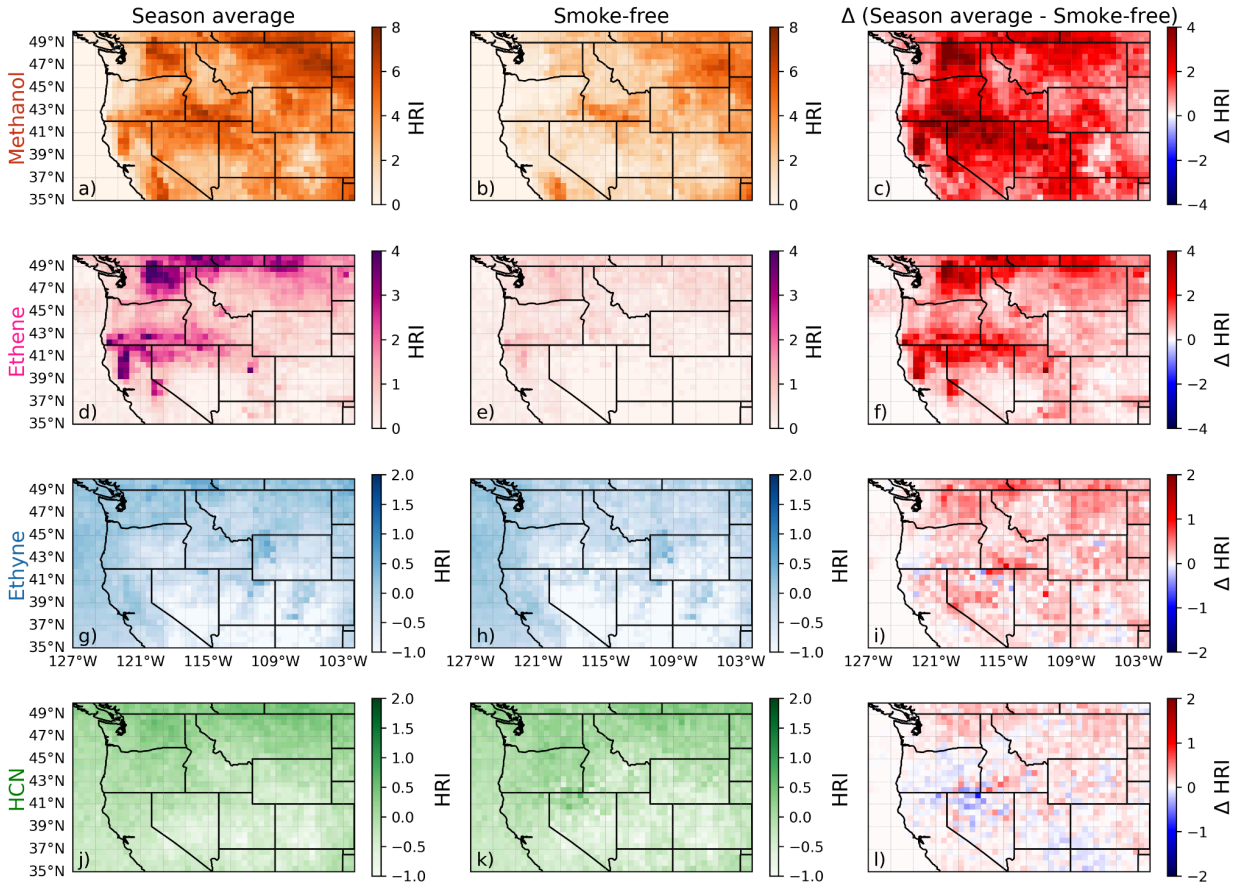


Figure S4. Spatial distribution of methanol, ethene, ethyne, and HCN spectral signals (HRIs) as measured by CrIS from July 24, 2018 - September 13, 2018. The left column shows the mean measured signal across the entire period, the middle column shows the mean for low/no-smoke periods, and the right column shows the difference. Data are plotted on a 0.5º × 0.5º grid and span the WE-CAN campaign timeframe.

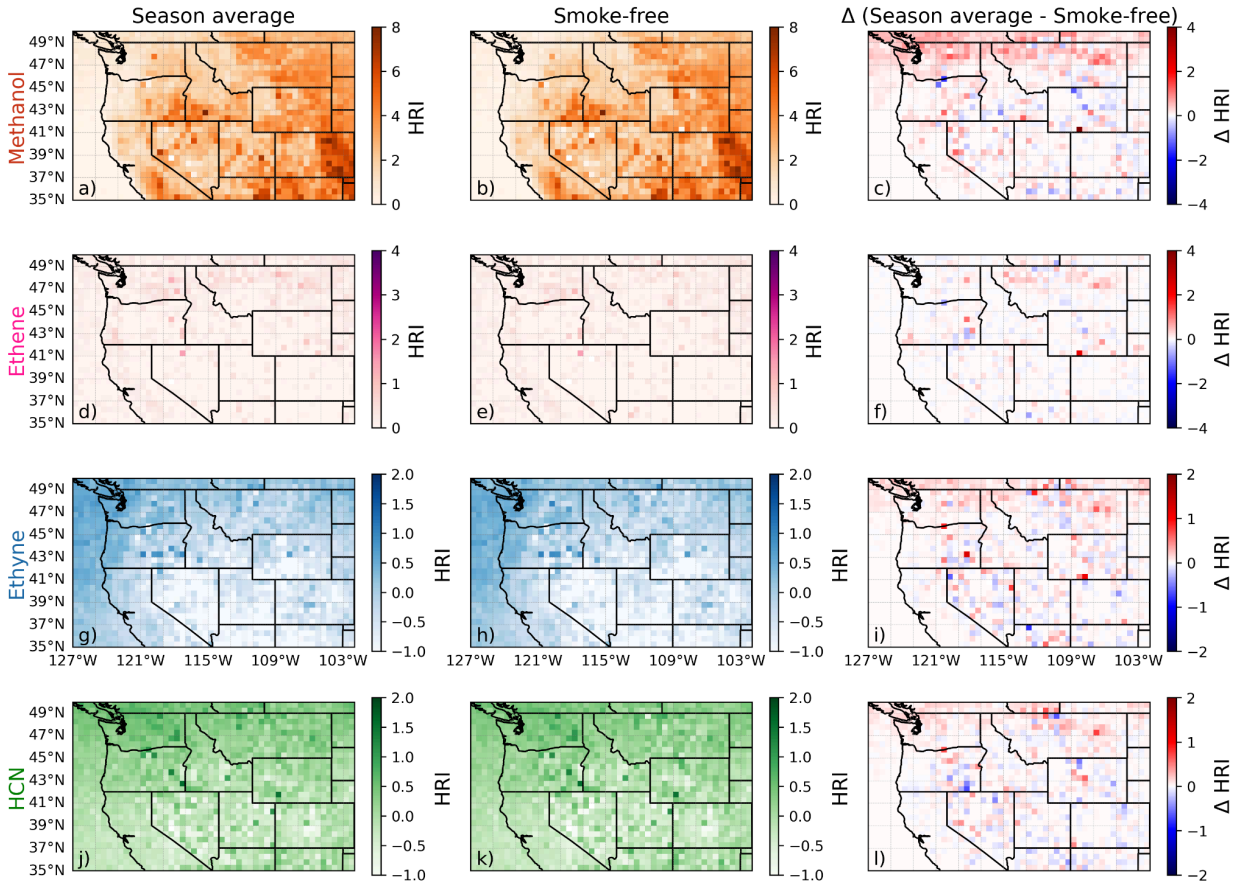


Figure S5. Spatial distribution of methanol, ethene, ethyne, and HCN spectral signals (HRIs) as measured by CrIS from July 26, 2019 - September 6, 2019. The left column shows the mean measured signal across the entire period, the middle column shows the mean for low/no-smoke periods, and the right column shows the difference. Data are plotted on a 0.5º × 0.5º grid and span the FIREX-AQ campaign timeframe.

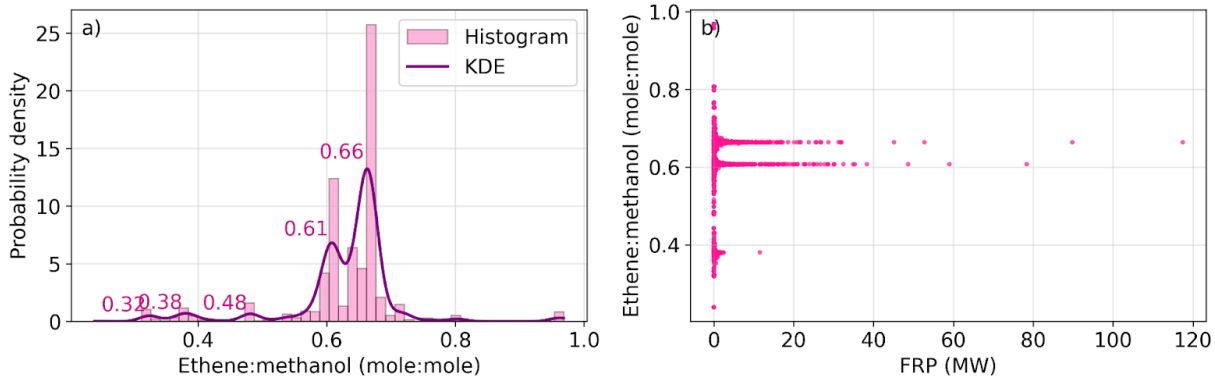


Figure S6. a) GFAS ethene:methanol emission ratio distribution for fires during 2018 and 2019 over the western U.S. The solid line shows the associated probability density based on kernel density estimation (KDE). b) The same data plotted as a function of GFAS FRP (MW).

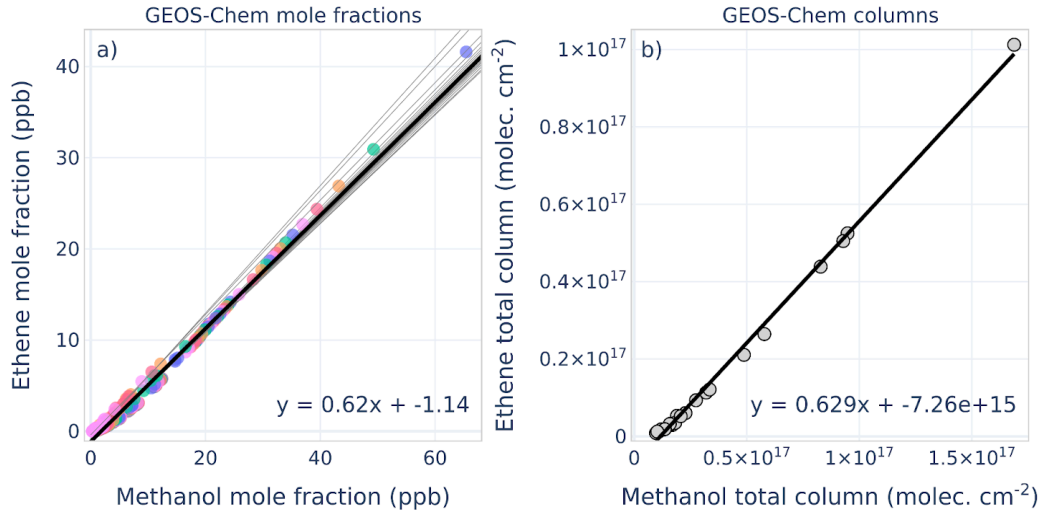
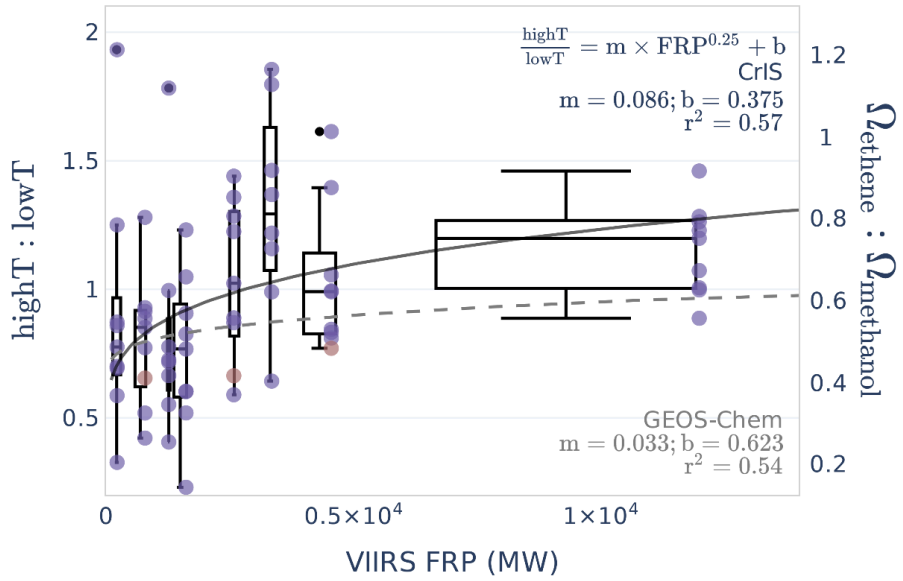


Figure S7. Pyrogenic ethene:methanol ratios are similar when computed using a) in-plume mole fraction and b) total column amounts. Data plotted are from the GEOS-Chem model for the Pole Creek Fire plume. Panel a) shows mole fraction regressions by pressure level from the surface to ~500 hPa (light grey lines) along with the regression across all pressure levels (black line). Panel b) shows a regression for total column values (surface to ~0.1 hPa). In both cases, the derived slopes are close to the molar ethene:methanol emission ratio used in the model for this fire (0.61).



S8. High-T:low-T pyrolysis ratio for each of these fire plumes (estimated from the CrIS ethene:methanol ratios as described in-text) as a function of VIIRS fire radiative power (FRP). Right axis shows the CrIS ethene:methanol ratios. Boxplots encompass the interquartile range (IQR) for each data bin; whiskers span datapoints within 1.5×IQR of the first and third quantiles. Boxplots are positioned at the bin-median FRPs with widths indicating the corresponding standard deviations. Regression coefficients inset reflect a fourth-root power-law relationship and are calculated using the median high-T:low-T ratio and FRP for each bin.