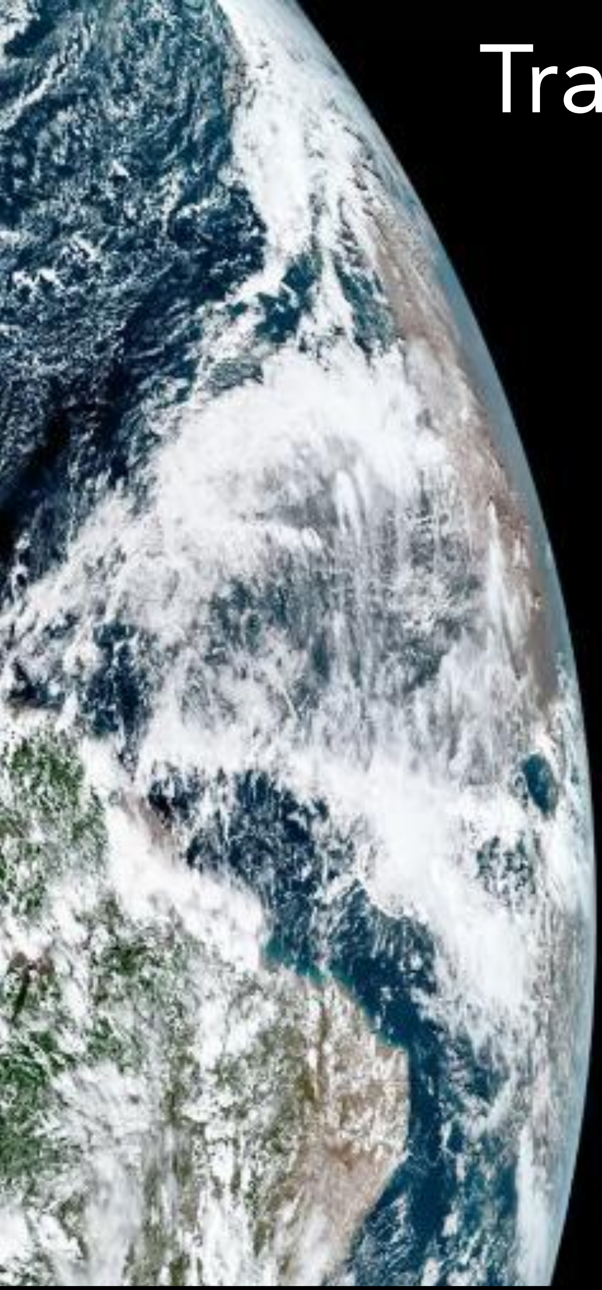

Trace Gas Opportunities for GXS and Synergy with ACX

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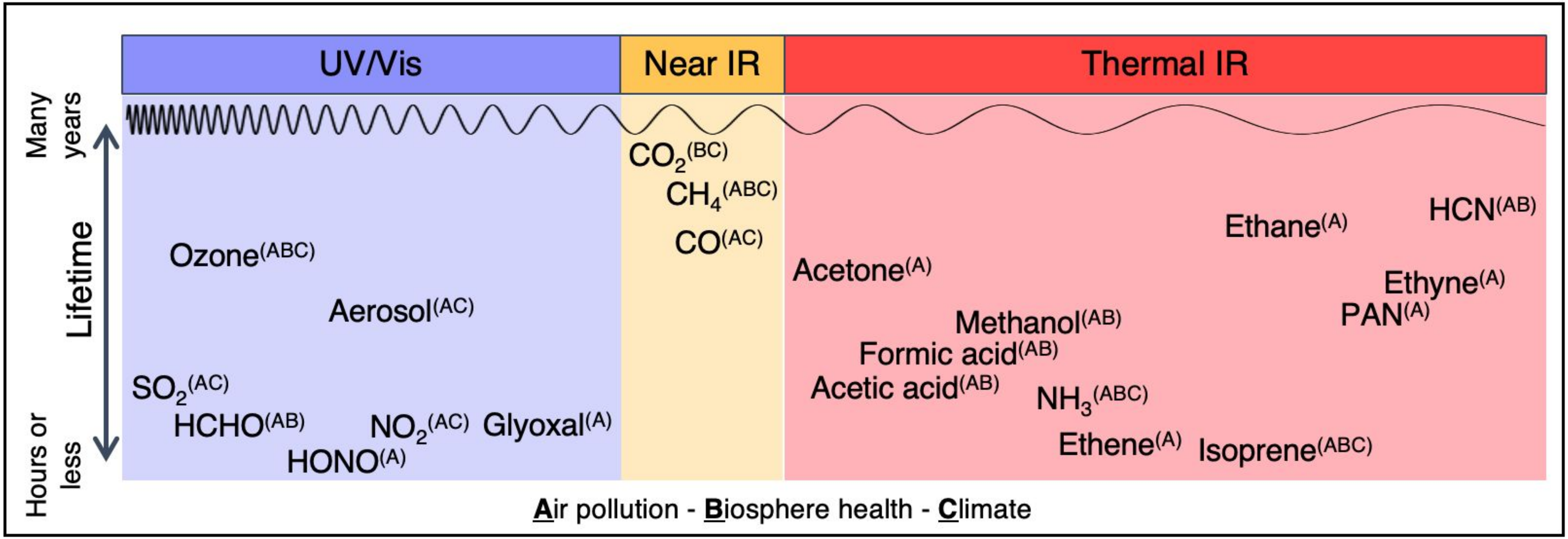
A range of key atmospheric chemicals are observable from space in the TIR

… and are not accessible in the UV/Vis



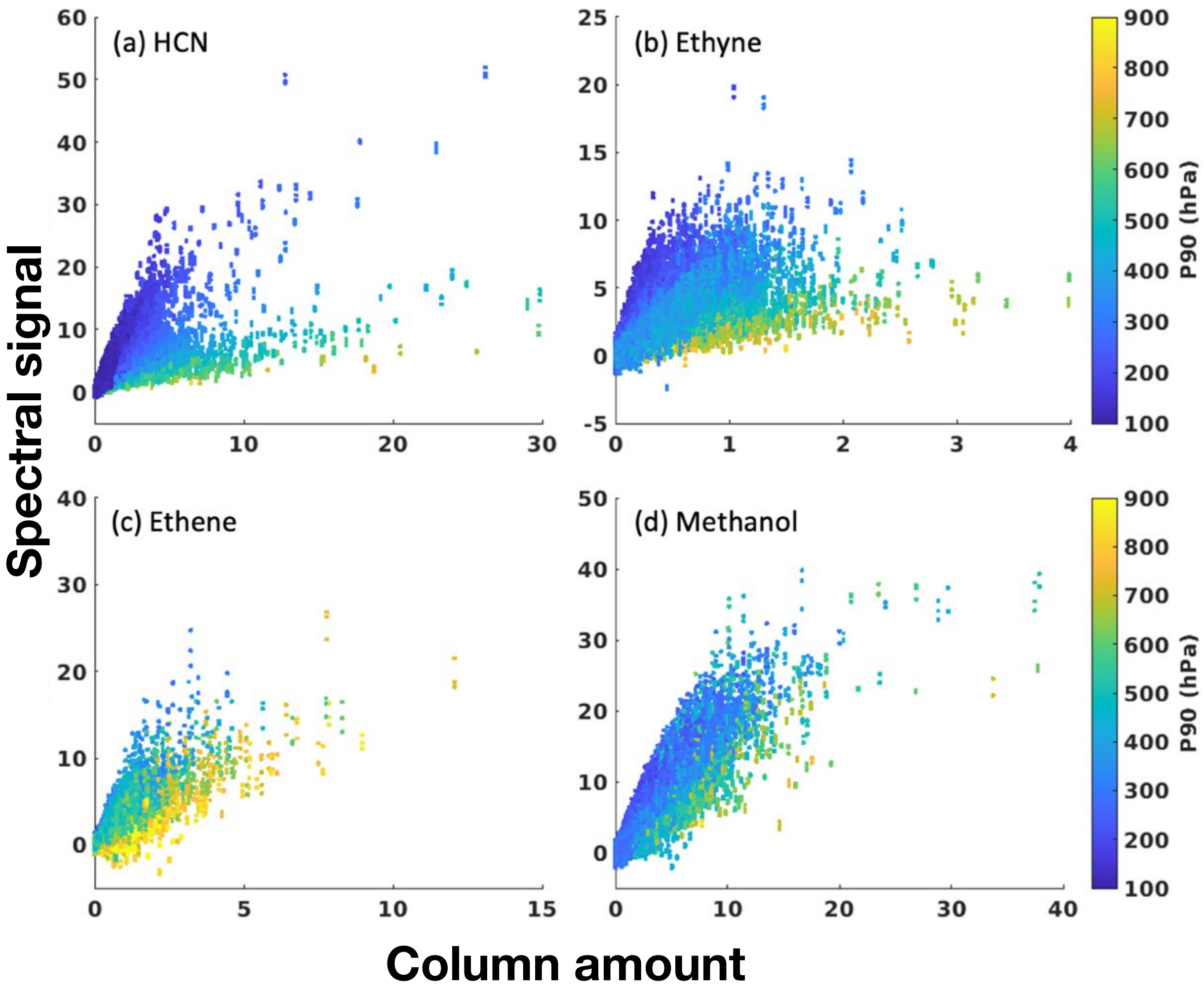
[Millet et al., *submitted*, 2024]

These TIR species are highly complementary with those that will be quantified by ACX

TIR measurements rely on the surface-atmosphere temperature difference

Detection efficiency increases with altitude

□ But TIR observations still provide sensitivity to the boundary layer!



This vertical dependence varies by compound

For example:

Methanol, ethene: ~2× effect

Ethyne: ~5×

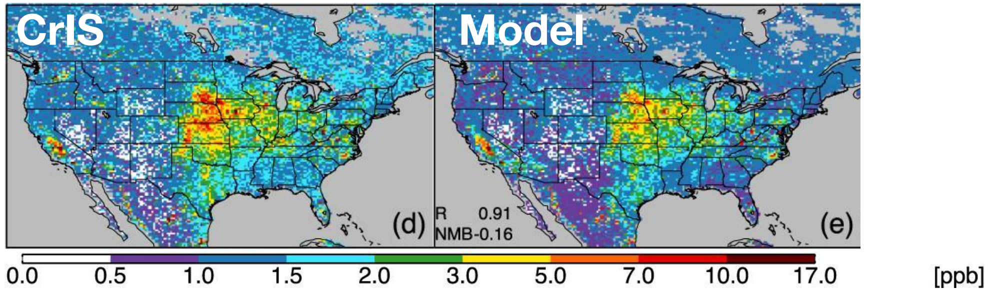
In practice this means that TIR retrievals require a constraint on the absorber's vertical distribution

Recent work with CrIS and IASI demonstrates value of TIR retrievals of VOCs, NH_3, PAN for air quality applications

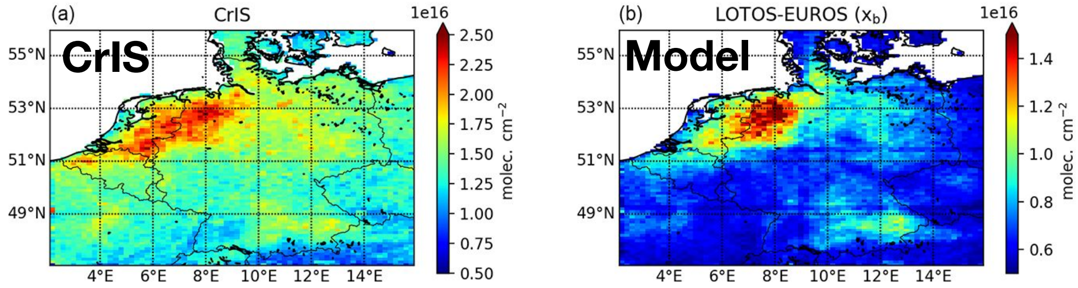
NH_3

Reactive N emissions caused ~23M years of life lost in 2013 due to resulting PM2.5

- Global NH_3 emissions now contribute more to PM2.5 than do NO_x emissions
- Marginal cost of abatement only 10% that of NO_x [Gu et al., *Science*, 2021]



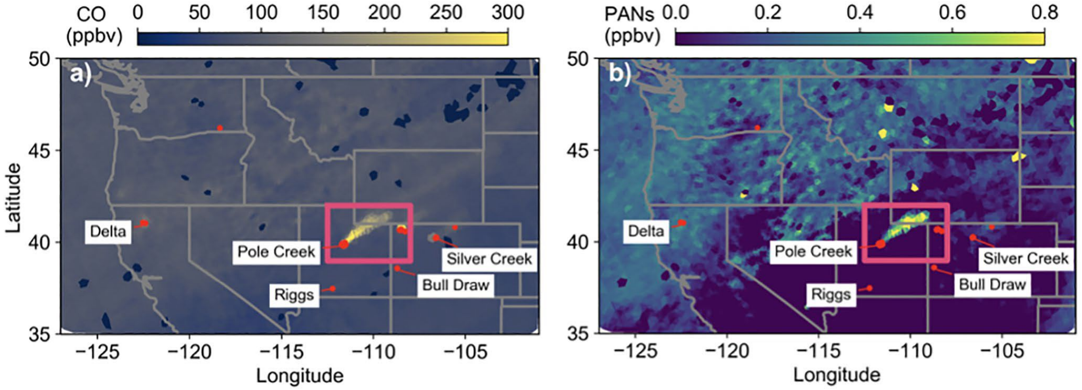
Inverse modeling of NH_3 emissions using CrIS [Cao et al., *ERL*, 2020]

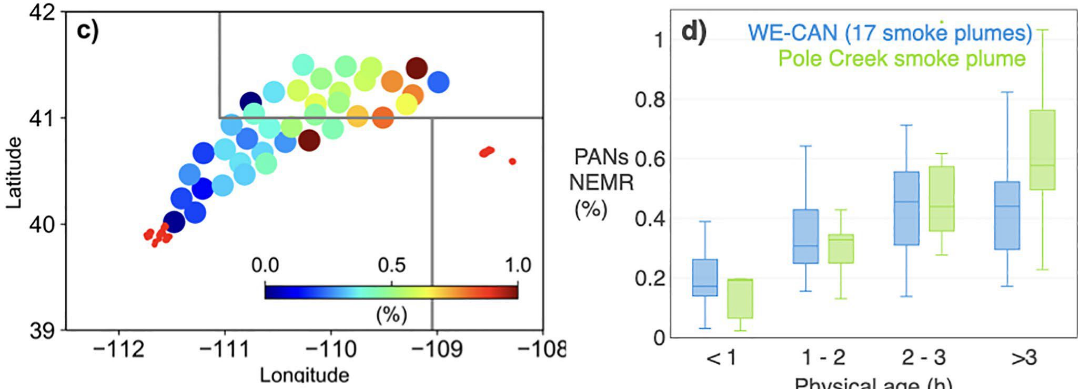


Assimilation of CrIS NH_3 to improve model simulations [van der Graaf et al., *ACP*, 2022]

PAN

Key NO_x reservoir, control on ozone



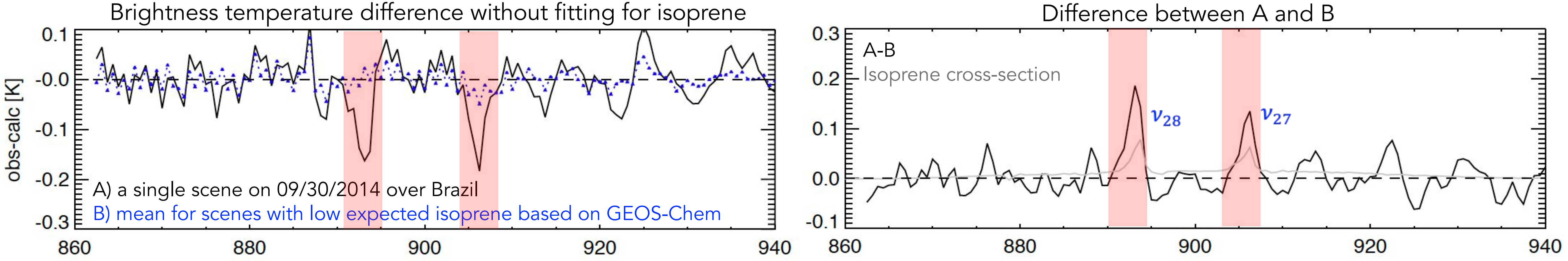


Using CrIS data to quantify PAN formation in fire plumes [Juncosa Calahorrano et al., *GRL*, *2021*]

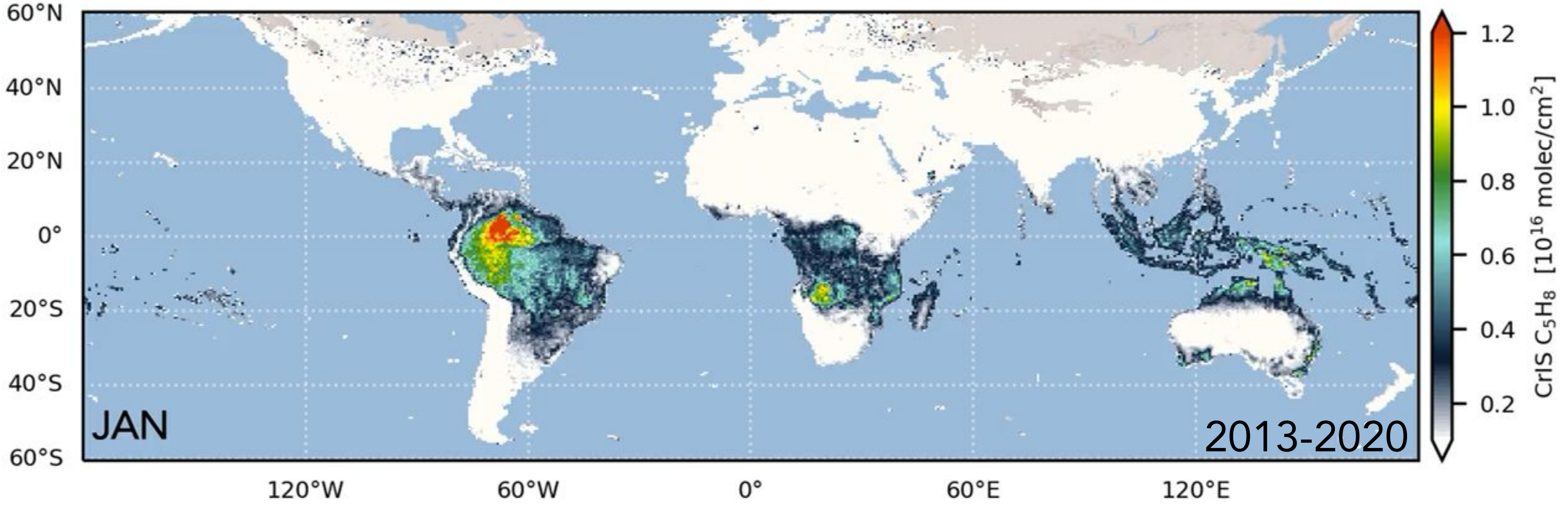
Measuring isoprene from space using CrIS

Example isoprene spectral signature in single CrIS spectrum

CrIS low noise provided enables first direct space-based measurement of isoprene



[Fu et al., *Nature Communications, 2019*]



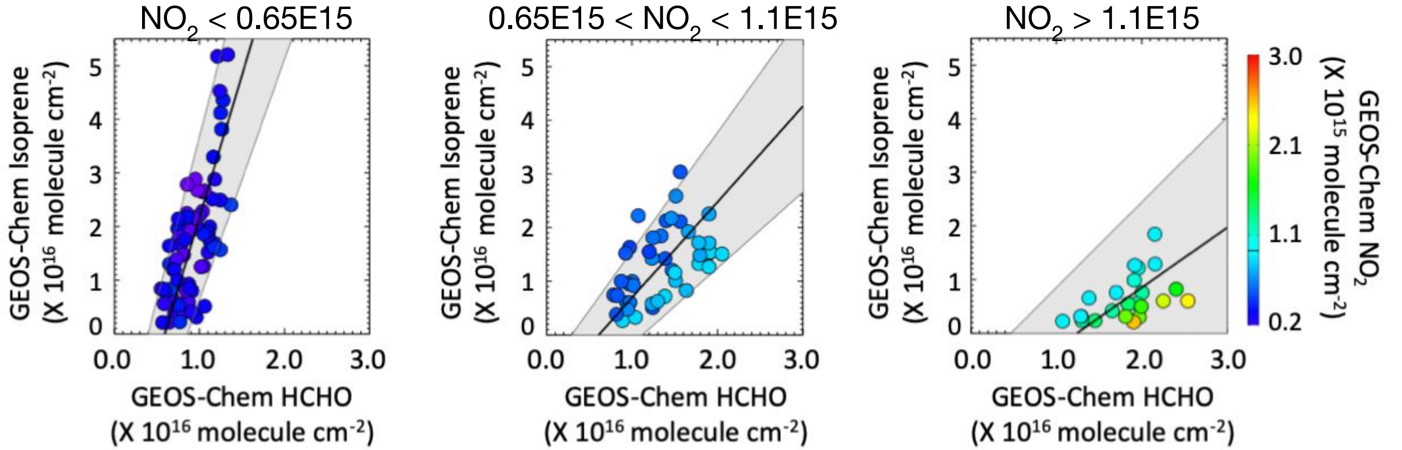
We are using a machine-learning framework to convert these spectral signals to isoprene column amounts through the CrIS record

□ Long-term global data record for studying ecosystem-atmosphere interactions & their chemical impacts

[Wells et al., *Nature* 2020]
[Wells et al., *JGR*, 2022]
[Shutter et al., *Science Advances*, in press]

Together, isoprene (GXS) and HCHO (ACX) measurements constrain isoprene emissions and atmospheric oxidation

GEOS-Chem isoprene:HCHO correlations over Amazonia binned by NO_2



[Fu et al., *Nature Communications*, 2019]

Decreasing NO_x

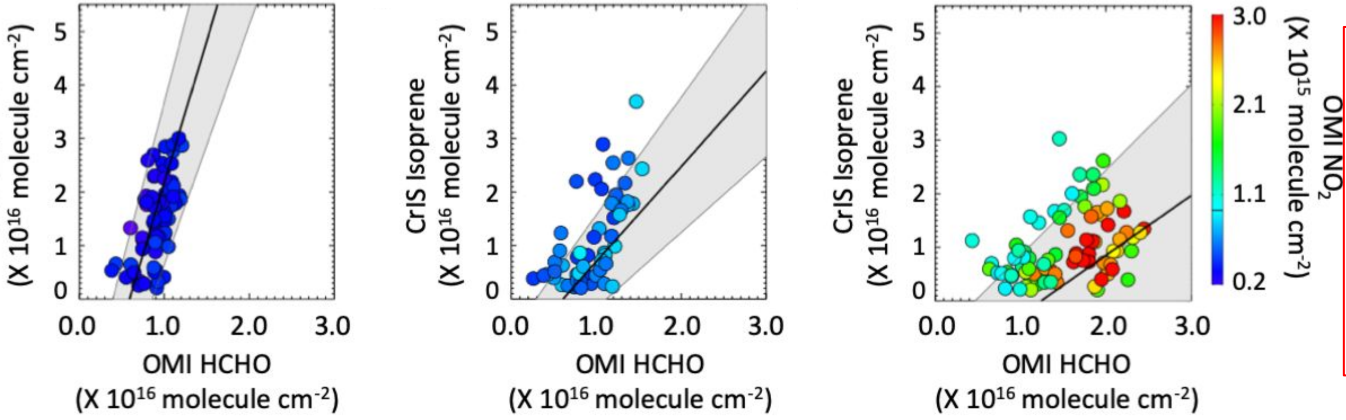
Isoprene:HCHO slope increases

Mainly due to suppressed OH, longer isoprene lifetime at low NO_x

Measured isoprene:HCHO correlations over Amazonia from CrIS+OMI, binned by OMI NO_2

Observed relationship at lowest NO_x consistent w. model

broadly supports OH recycling/ suppression in current mechanism

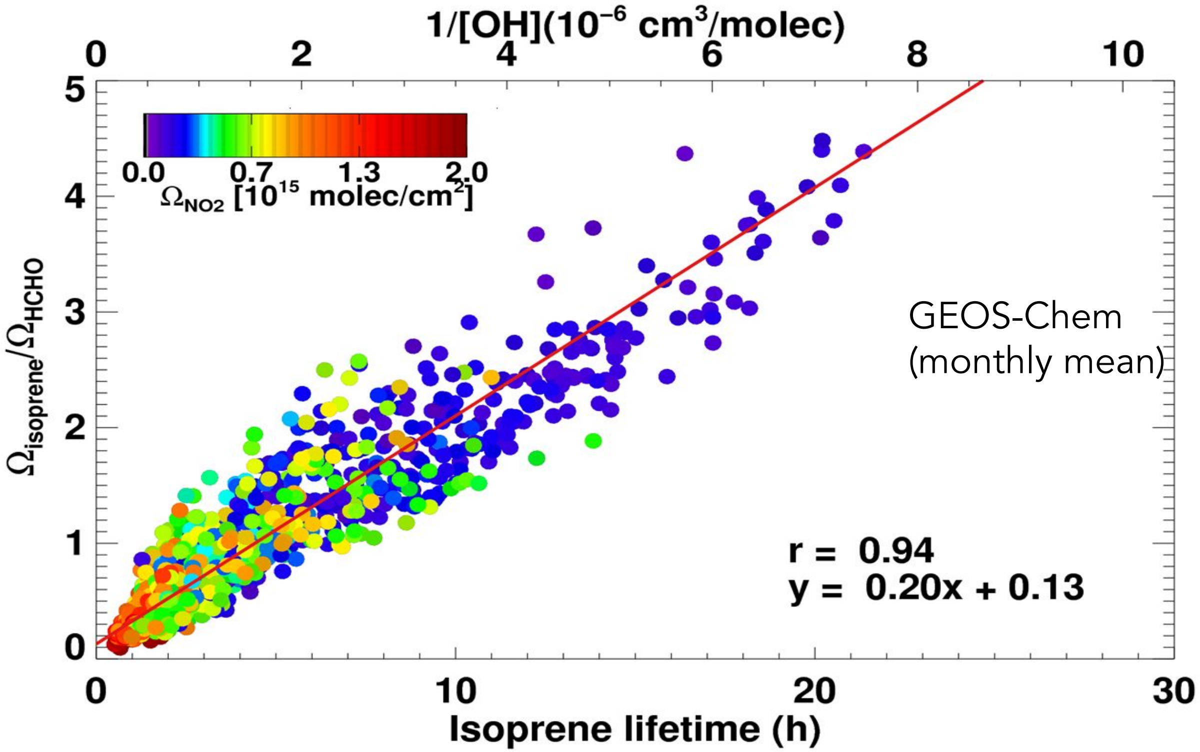


Some scatter above model relationship at higher NO_x

but no suggestion of underestimated OH recycling

Assessing atmospheric OH regimes from space

The isoprene:HCHO column ratio scales closely with OH



Isoprene, HCHO both related to isoprene emission

But with differing sensitivity to OH

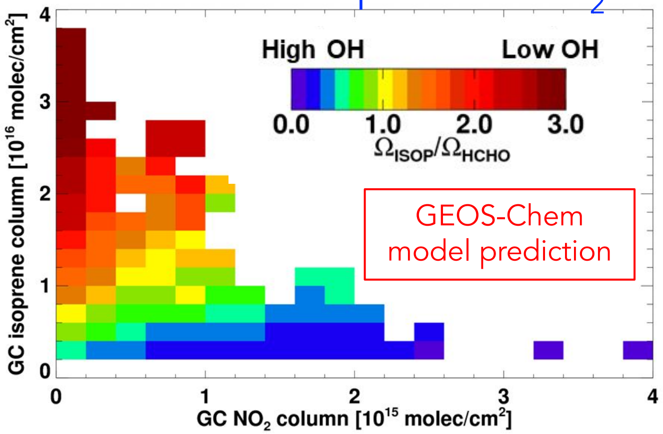
Complementary information on isoprene sources and sinks

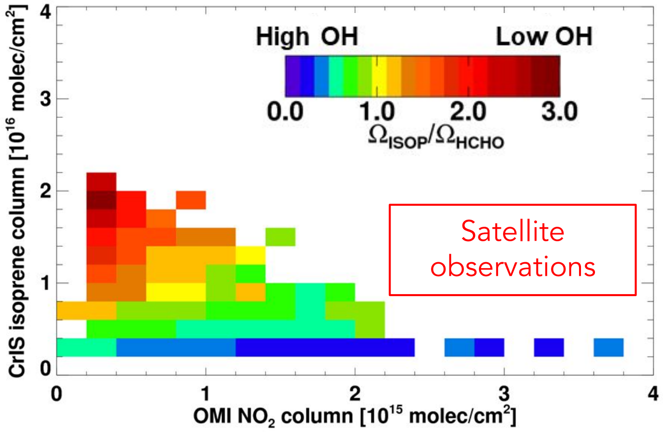
Space-based constraint on atmospheric oxidation over biogenic source regions

[Wells et al., *Nature*, 2020]

This allows us to test understanding of isoprene chemistry and its effects on OH

Isoprene lifetime & OH as a function of isoprene & NO_2



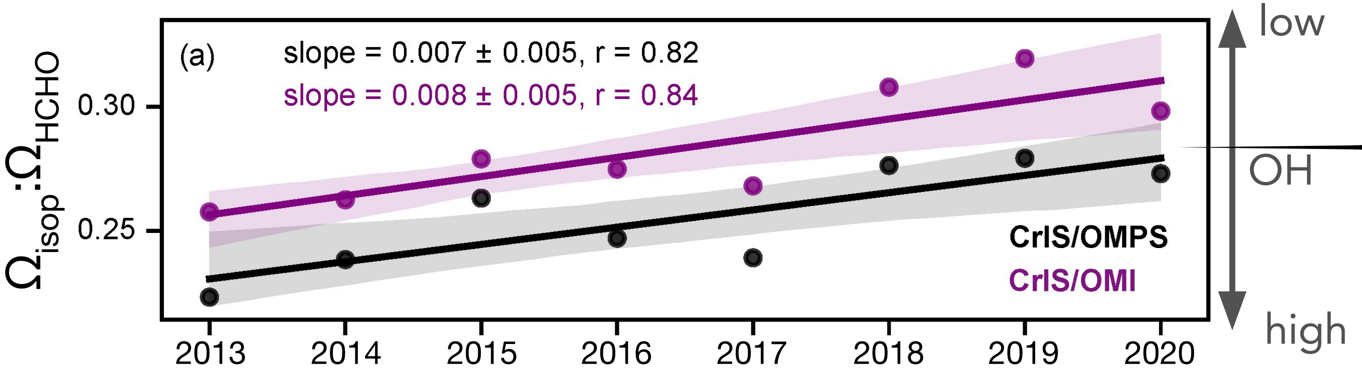


OH suppressed with increasing isoprene and decreasing NOx

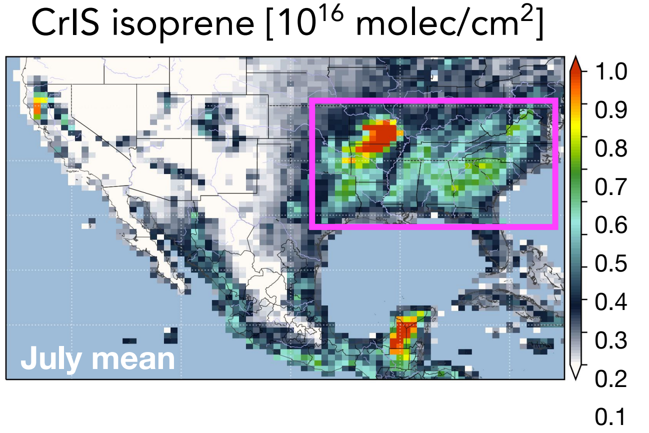
These observed shifts in chemical regimes are similar to model predictions

Argues against significant missing OH recycling

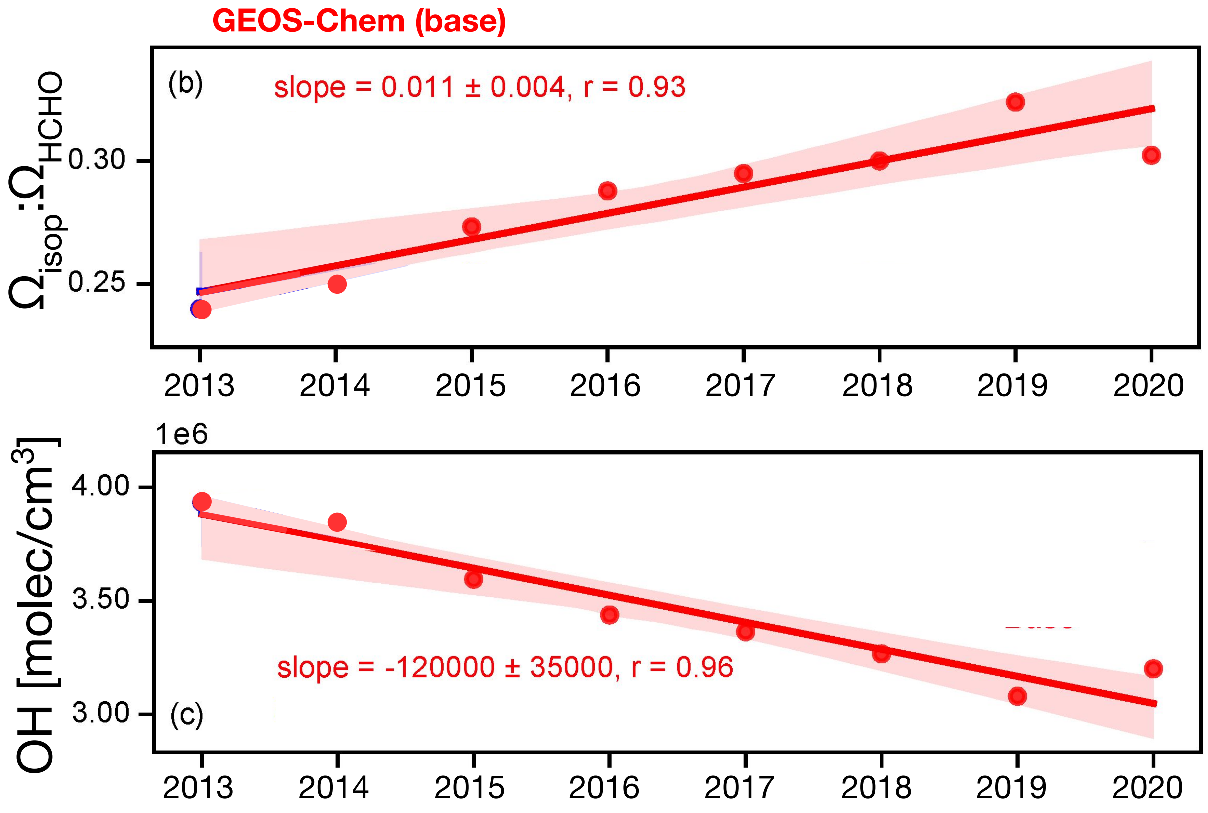
Mapping OH trends over the Southeast US



Satellite measurements reveal multi-year Ω_{isop}: Ω_{HCHO} trend

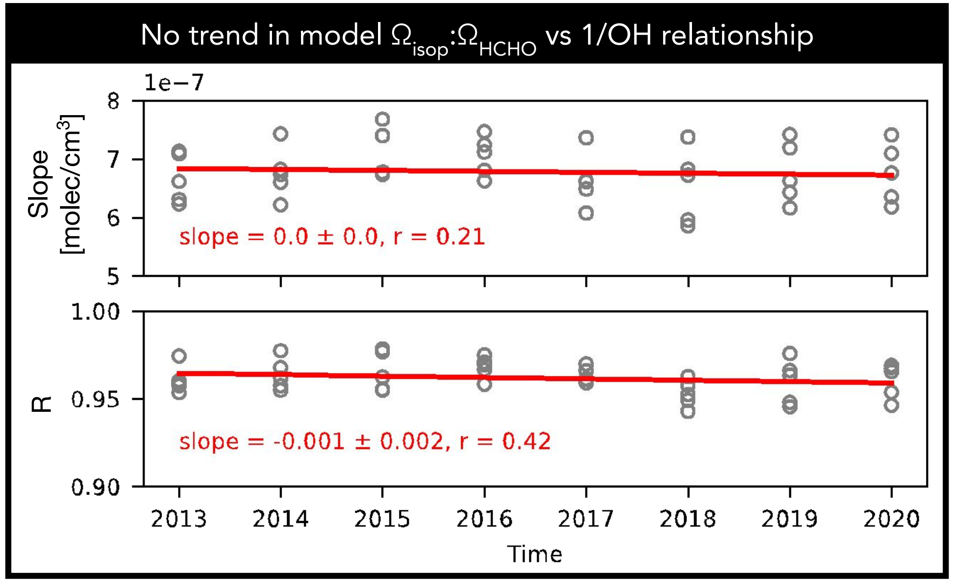


[Shutter et al., *Science Advances*, in press]

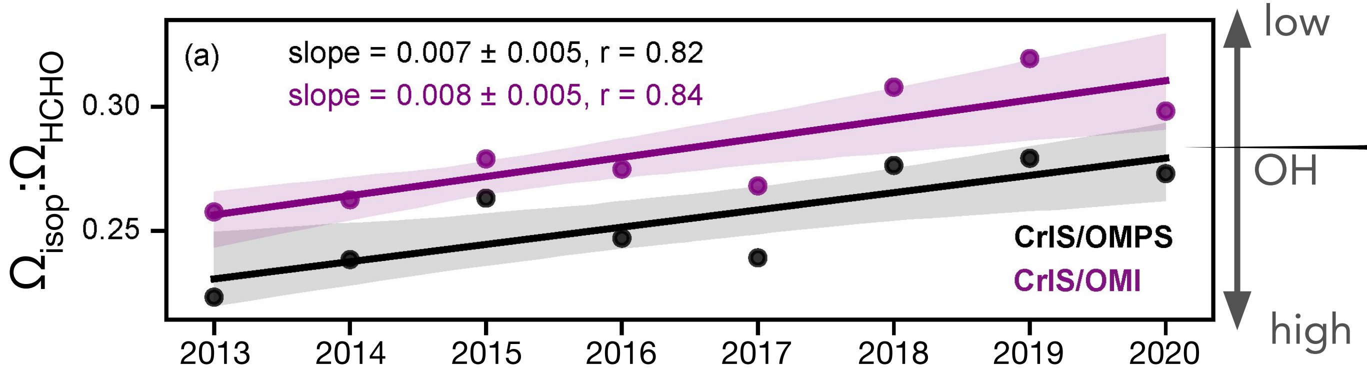


- *Corroborating the model-predicted trend…*

… which is due to changes in OH, not in the Ω_{isop}:Ω_{HCHO} vs. 1/OH relationship

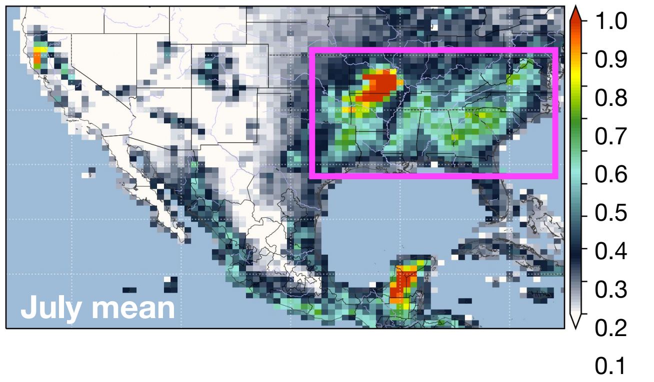


Mapping OH trends over the Southeast US

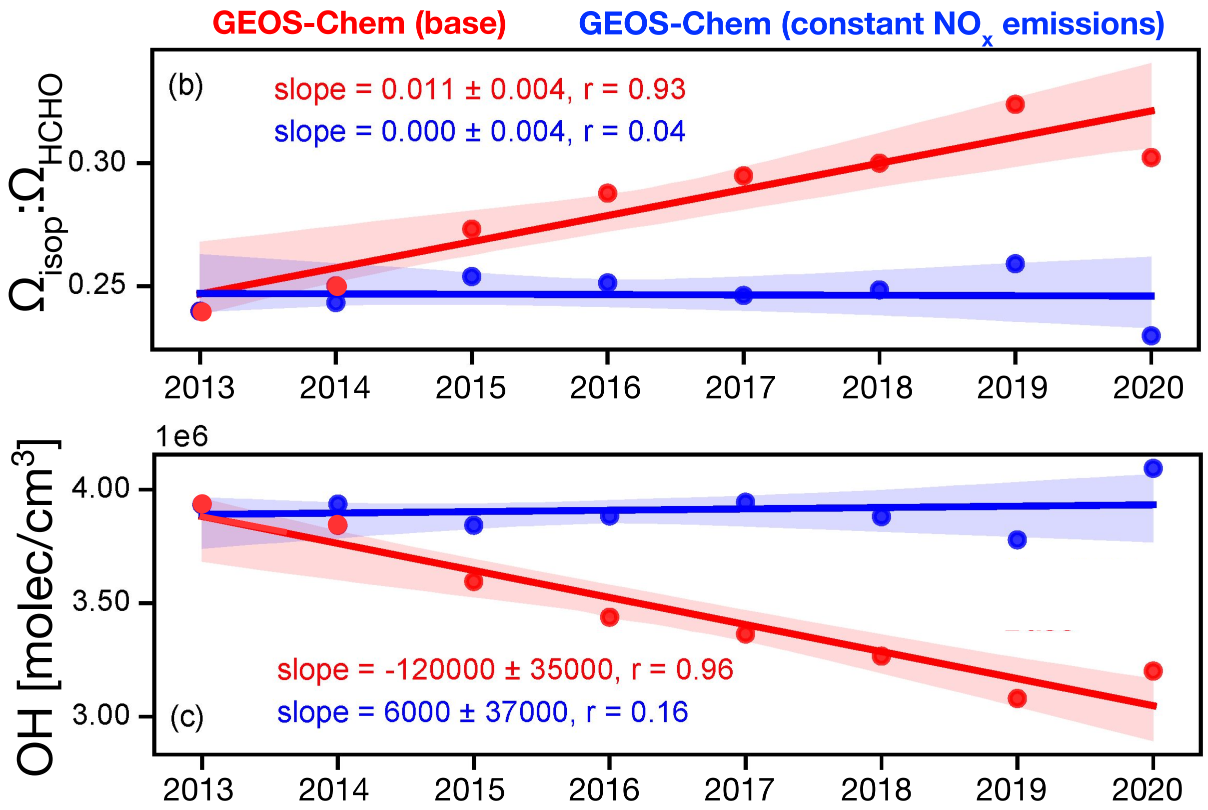


Satellite measurements reveal multi-year Ω_{isop}:Ω_{HCHO} trend

CrIS isoprene [10^{16} molec/cm^2]



[Shutter et al., *Science Advances*, in press]



- *Corroborating the model-predicted trend…*

… which is due to changes in OH, not in the $\Omega_{isop}:\Omega_{HCHO}$ vs. 1/OH relationship

Model OH trend driven by decreasing anthropogenic NO$_x$ source

- *Total OH decline:* 8×10^5 molecules cm^{-3} (21% of summer daytime peak)

OH decrease diagnosed from space

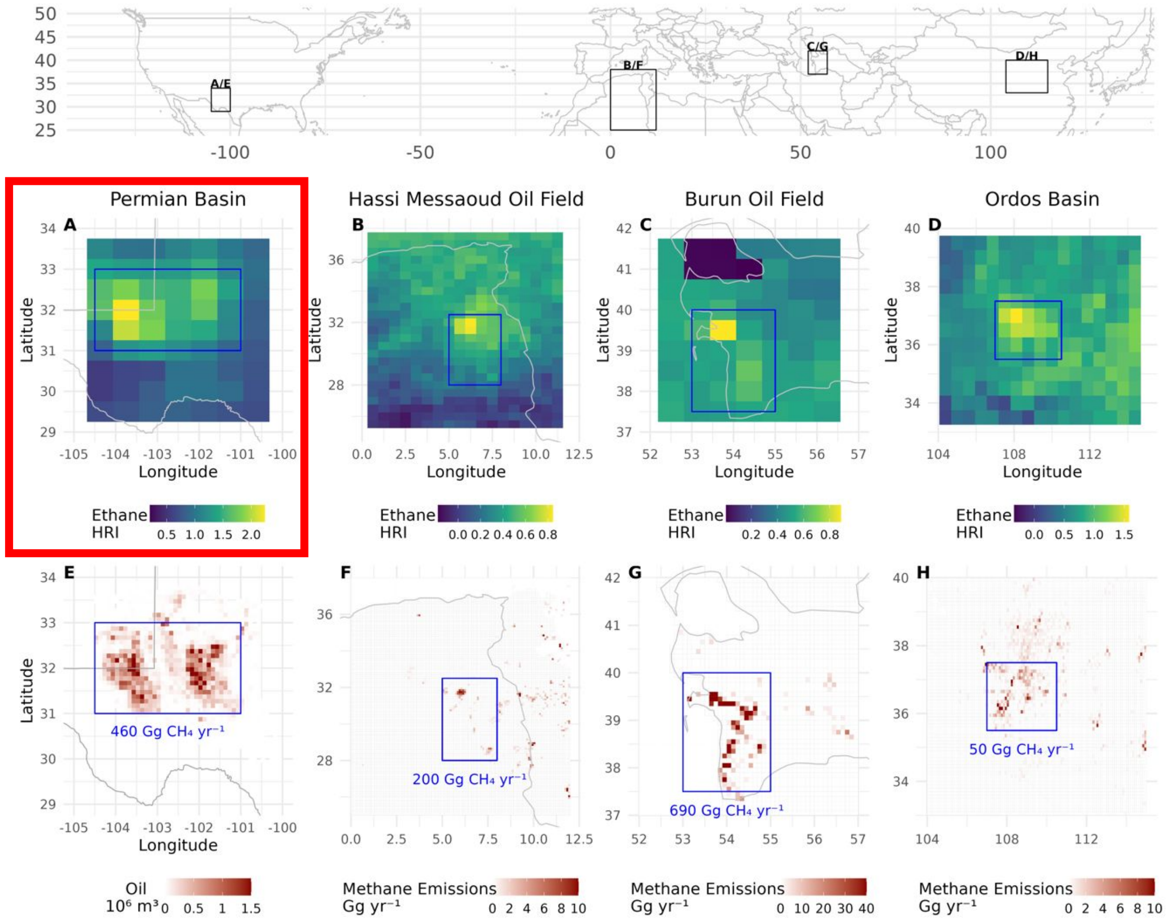
- Linked to changes in regional O_3, NO_x, SO_2, SOA

[Simon et al., 2015, Li et al., 2018; Blanchard 2016; Marais et al., 2017; Zheng et al. 2020; Boris et al., 2021]

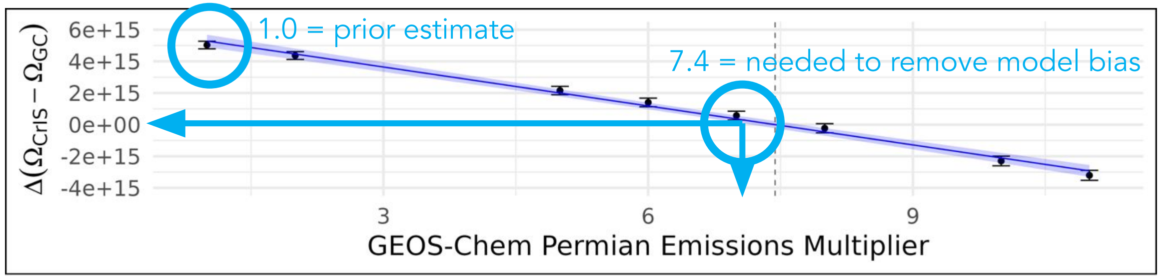
- Shifting chemical climatology of SE US

Space-based observations of tropospheric ethane map fossil fuel emissions

[Brewer et al., in revision]



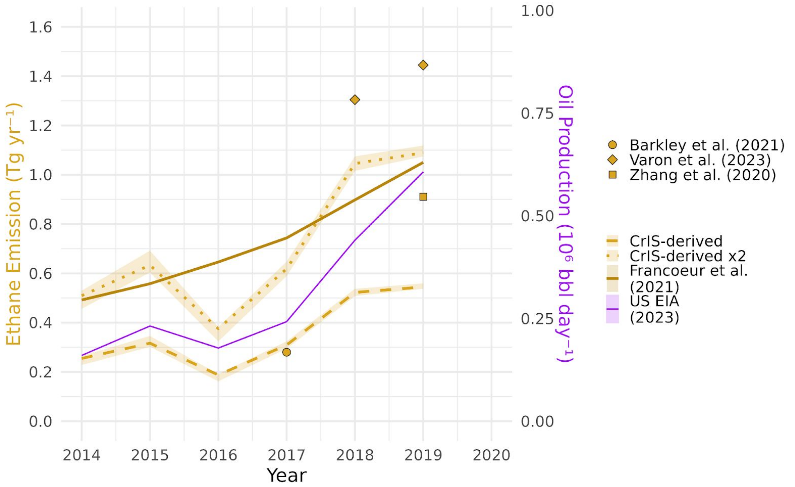
CrIS measurements show that the Permian fossil fuel production basin in Texas and New Mexico stands out globally with the largest persistent ethane enhancements on the planet



Sensitivity simulations show that the basin-wide ethane flux predicted by GEOS-Chem needs to be scaled up 7× to match CrIS observations

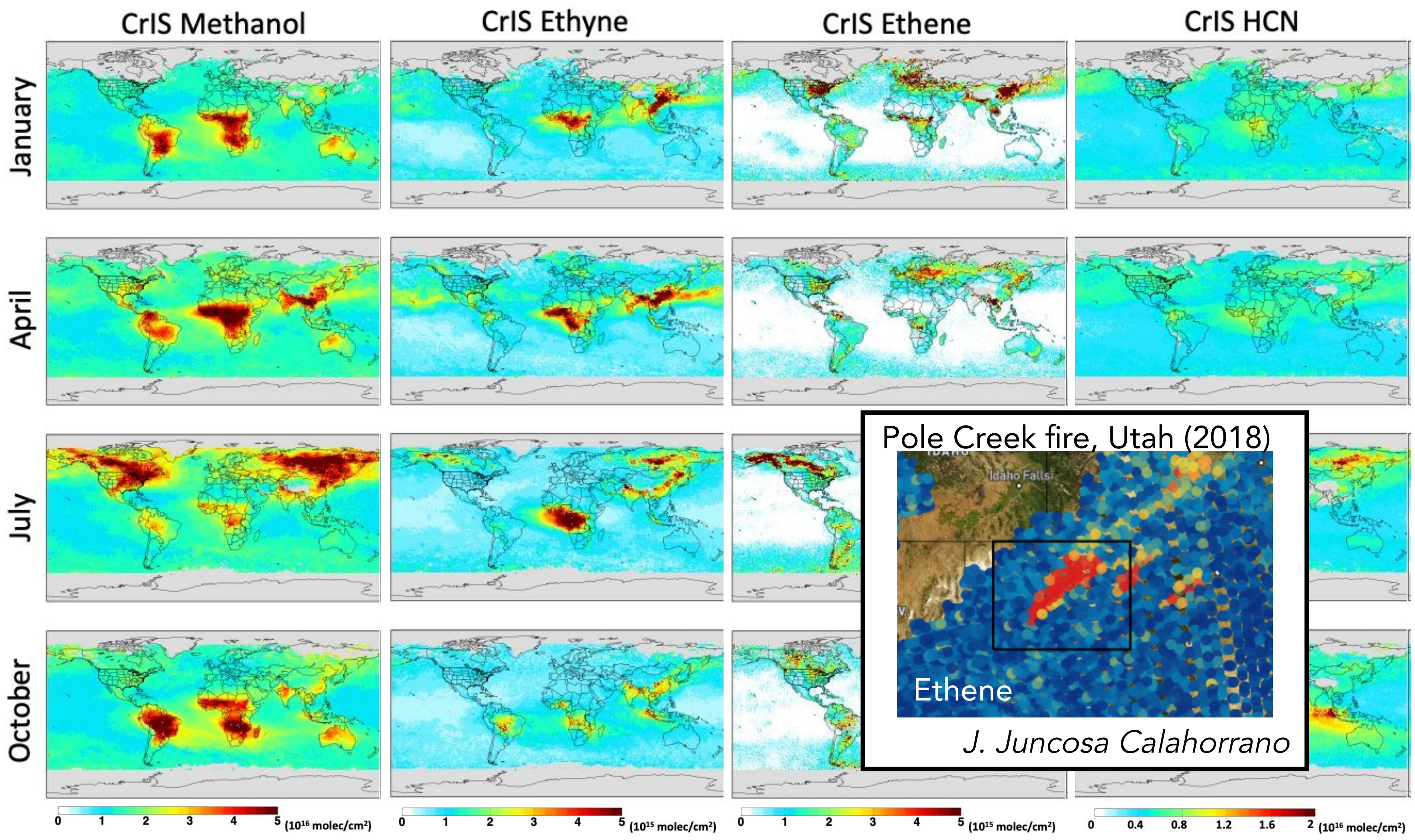
Permian basin alone then accounts for at least 4-7% of total global fossil-fuel ethane source

CrIS-derived emissions track oil/gas production rates and new FOG inventory



GXS observations would also enable measurements for a suite of other VOCs

Tracers for understanding biogenic, pyrogenic, anthropogenic emissions and changes over time

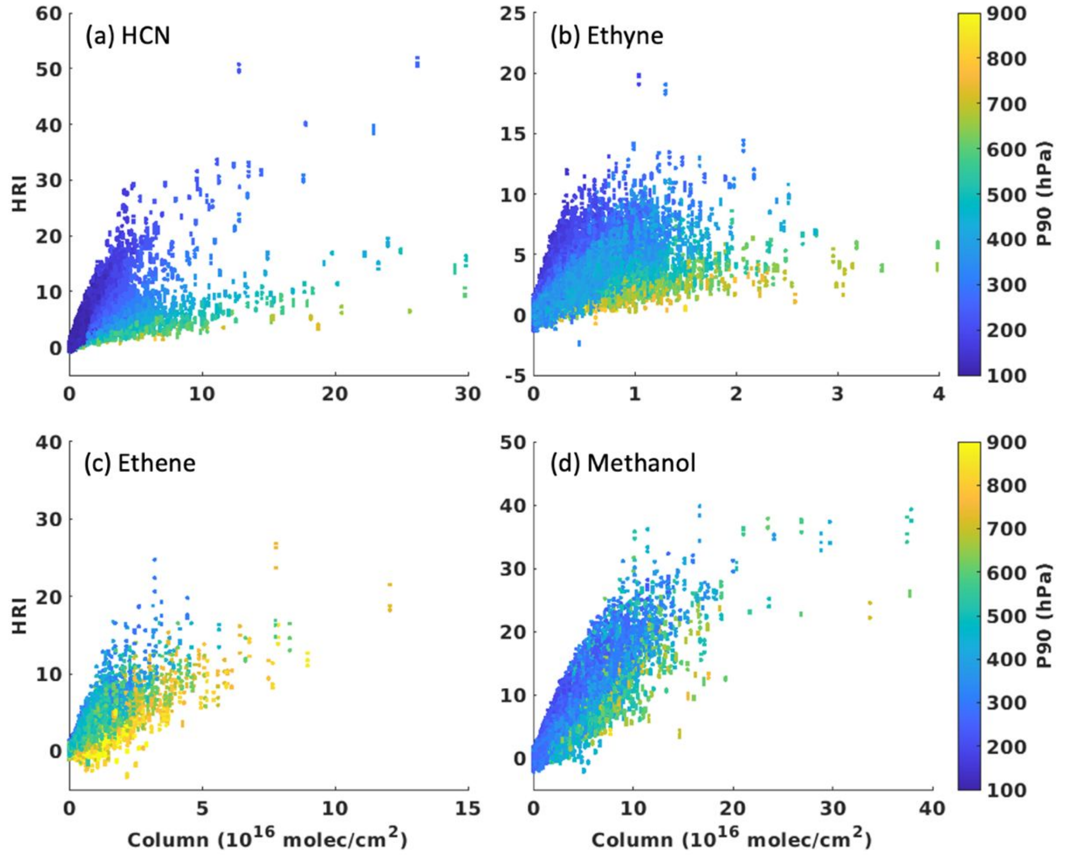


[Wells et al., *to be submitted*, 2024]

Extra

Vertical sensitivity of TIR measurement

[Wells et al., *to be submitted*, 2024]



ANN training sets used in the ROCRv2 VOC retrievals, illustrating the HRI-column relationship and the vertical dependence of detection sensitivity. Data are plotted for (a) HCN, (b) ethyne, (c) ethene, and (d) methanol, and are shaded by P_{90}, the atmospheric pressure below which 90% of the VOC column resides.

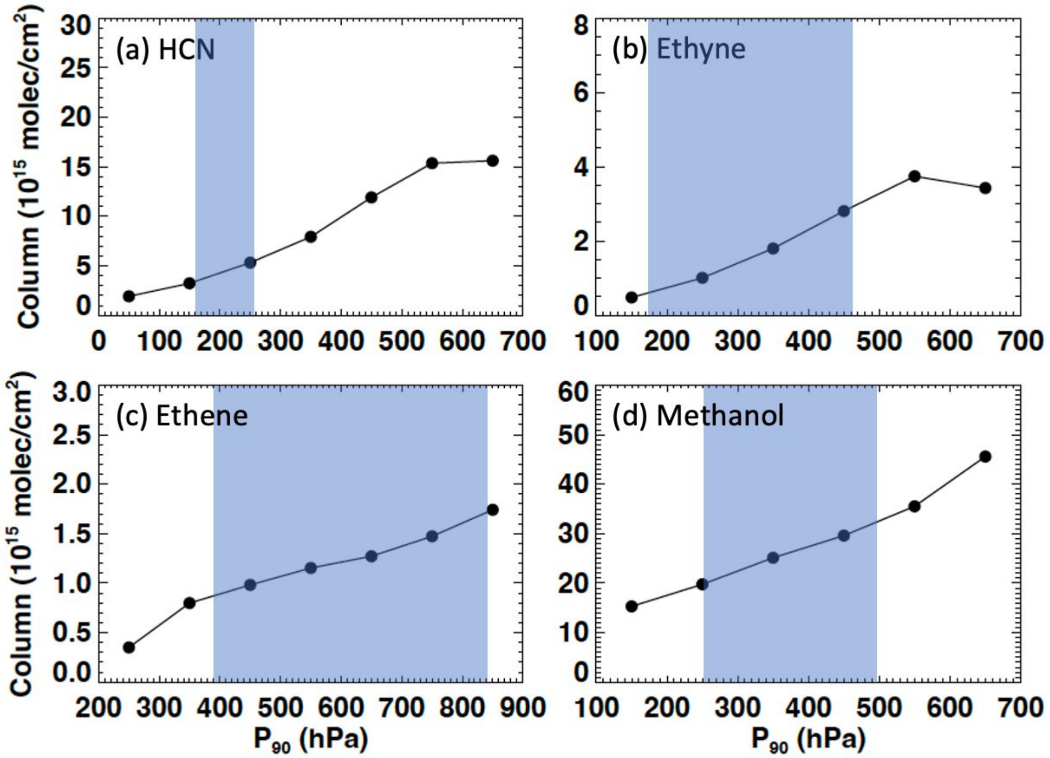
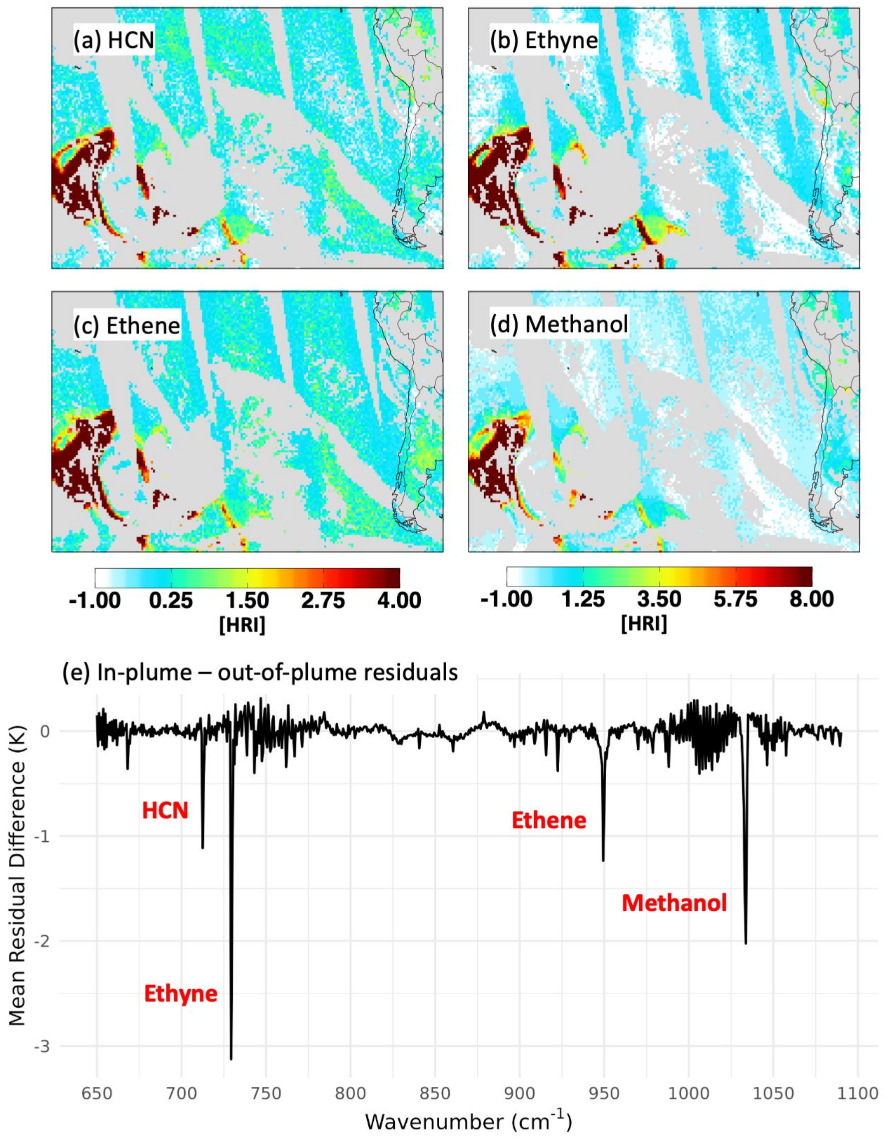
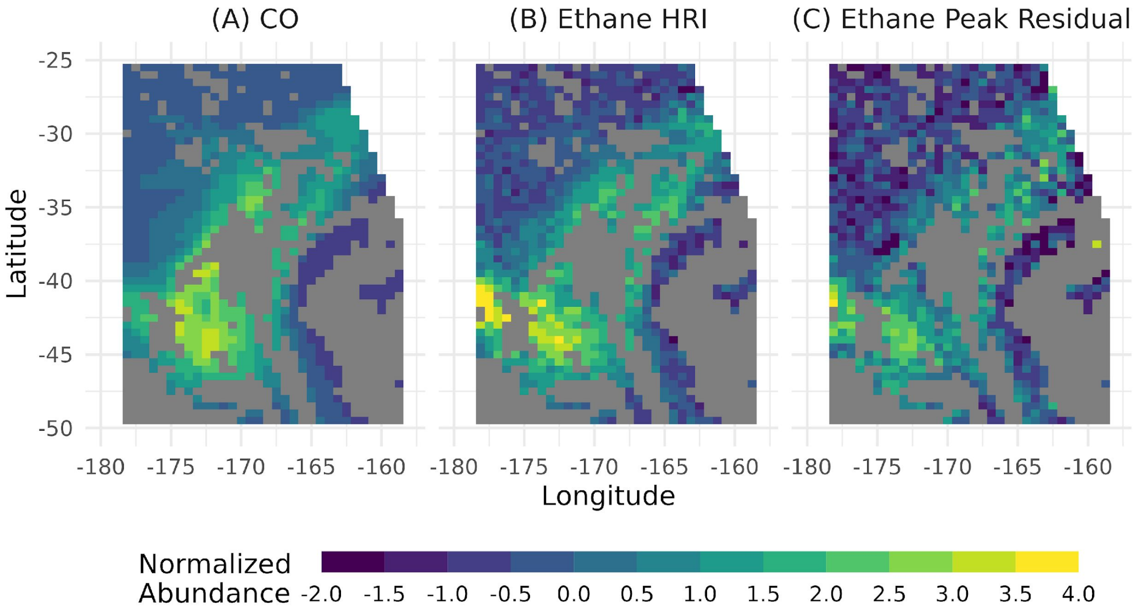


Illustration of CrIS retrieval sensitivity to VOC vertical profile assumptions. Shown are the CrIS-retrieved column enhancements for October 2019 over Amazonia (mapped in Fig. S3) as derived using each of the globally-fixed P_{90} values employed in the ROCRv2 ANN. Shading indicates the range in P_{90} values encountered over this region based on GEOS-Chem predictions for each species.

VOC spectral signals seen from space by CrIS



[Wells et al., *to be submitted*, 2024]



Spectral detection of ethane in CrIS radiances. Plotted are (a) CrIS CO Columns [43], (b) ethane HRIs, and (c) ethane brightness temperature differences for a fire plume over the South Pacific on January 2, 2020. All quantities are normalized to their largest value and screened for clouds using the 900 cm^{-1} brightness/surface-skin temperature difference[28]. Ethane HRIs and brightness temperature differences are computed as described in-text.

[Brewer et al., *in revision*, 2024]