

VOC and Methane Emissions from U.S. Oil and Natural Gas Production Barbara Dix¹, Alexander C. Bradley^{1,2}, Fergus Mackenzie³, and Joost de Gouw^{1,2} 1 CIRES, 2 Department of Chemistry, 3 BlueSky Resources, Boulder, CO

https://sites.google.com/view/de-gouw-lab

Background

- Oil and natural gas (O&G) production is associated with NOx, VOC and methane emissions.
- NOx and VOCs lead to ozone formation, affecting local and regional air quality. Methane is a potent greenhouse gas.
- These emissions are generally not well monitored. Satellite observations can help.

Instruments & Methods

- We study emissions from O&G production satellite observations of NO₂, formaldehyde (HCHO) and methane (CH₄) TROPOspheric Monitoring the from Instrument (TROPOMI).
- TROPOMI provides daily global coverage at ~1:30pm local time, with a footprint of 3.5x5.5km² for NO2 and HCHO and 7x5.5 km² for CH₄.
- Divergence Technique: Horizontal gradients in background corrected satellite vertical columns, Vcorr, multiplied with horizontal boundary layer wind fields, u, provide source location and strength of underlying emissions, E: $E = \mathbf{u} \cdot \nabla V_{corr}$
- Tensorflow machine learning (ML) deep transfer learning model framework was used to train an annualized model on 2018-2022 TROPOMI data, then retrained on monthly data subsets. Target data was GOSAT proxy retrieval measurements (see schematic below).



References

(1) Barbara Dix, Meng Li, Esther Roosenbrand, Colby Francoeur, Steven S. Brown, Jessica B. Gilman, Thomas F. Hanisco, Frank Keutsch, Abigail Koss, Brian M. Lerner, Jeff Peischl, James M. Roberts, Thomas B. Ryerson, Jason M. St. Clair, Patrick R. Veres, Carsten Warneke, Robert J. Wild, Glenn M. Wolfe, Bin Yuan, J. Pepijn Veefkind, Pieternel F. Levelt, Brian C. McDonald, and Joost de Gouw: Sources of Formaldehyde in U.S. Oil and Gas Production **Regions,** ACS Earth and Space Chemistry (2023).

Acknowledgements

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Figure 1: Top: 2018/19 summer (JJA) averages of a) HCHO and b) NO₂ VCDs, and c) O&G production. Bottom: Results of spatial correlation analysis. White points mark correlation maxima; arrows indicate ECMWF wind vectors at 100 m above ground

WRF-Chem sensitivity studies show:



Figure 2: HCHO VCDs over the Permian basin for 10 July to 16 August, 2018, a) observed by TROPOMI, b)-d) simulated by WRF-Chem. Numbers denote average VCD for the area marked by a black box. Acknowledgements: WRF-Chem model data: Meng Li and Brian McDonald; FOG data: Colby Francoeur and Brian McDonald.

Analysis of aircraft in-situ data shows:

- emissions (Fig. 3).
- fractions.



Steven Brown, Robert Wild.

 \rightarrow About 96% of observed HCHO during March/April is the product of VOC oxidation. \rightarrow Total primary O&G HCHO emissions are between 6-1500 kg/h per basin.

Formaldehyde and VOC Emissions (1)

TROPOMI observations and spatial correlation analysis show:

• Summer HCHO and NO₂ Vertical Column Densities (VCDs) over the Permian basin, one of the largest O&G basins, are enhanced over areas with high production (Fig. 1a-c). • A strong correlation between observed gases and production locations (Fig. 1d/e). • A tighter spatial correlation of production with NO₂ (Fig. 1f).

 \rightarrow Consistency with O&G NO_x emissions and secondary HCHO formation from O&G VOCs.

• HCHO simulated with Fuel-based Oil and Gas (FOG) inventory NO_x and VOC emissions are in good agreement with TROPOMI (Fig. 2a/b).

• O&G precursor VOCs lead to HCHO formation, as do other anthropogenic VOCS. The HCHO yield is highly sensitive to NO_x (Fig. 2d/e).

 $\rightarrow O\&G VOC$ and NO_x emissions are equally important to VOC oxidation chemistry.

• Observed HCHO correlates well with PAN - here a tracer for VOC oxidation - and only sporadically, in fresh emission plumes, with NOx - here a tracer for primary O&G

• Multi-variate fitting of NO_x and PAN to HCHO provides primary and secondary HCHO

• The NOx fit factor can be used to scale FOG NO_x emissions to estimate primary HCHO emissions from O&G production (Fig. 4).





Figure 4: Estimate of O&G primary HCHO emissions for SONGNEX sampled basins. Acknowledgements: FOG data: Colby Francoeur and Brian McDonald.

Methane Emissions in the Denver-Julesburg basin - a feasibility study

Study questions:

- Can we derive methane emissions for the Denver-Julesburg basin using TROPOMI CH4 observations and the divergence technique?
- How are derived emissions impacted by the known ground-albedo bias in TROPOMI CH4 VCDs and can we correct for that?
- Can we separate O&G emission from other methane sources by their seasonal signal?

Sensitivity study on deriving methane emissions for the Permian basin (strong signals/high data coverage) shows:

- Derived Emissions depend on boundary layer wind speeds (Fig. 4), which should not happen in theory.
- At low wind speeds, methane pools around sources, creating a constant offset not picked up by the divergence calculations.
- Flux Divergence results are less wind speed dependent and might account for offsets, but cause bias in source locations (Figs. 4 and 5).



<u>Development of seasonal methane albedo correction</u> is motivated by:

- CH₄ columns correlate with surface albedo in the operational retrieval. Existing corrections are built on annual or longer timescales.
- Deconvoluting seasonal from non-seasonal oil & gas methane emissions could help build more accurate inventories.

<u>Application of seasonal methane albedo correction:</u>

- Is based on trained machine learning (ML) model with TROPOMI and GOSAT data
- Provides best correction of seasonal albedo bias compared to other correction methods (Fig. 6). Monthly Pearson Values for Different Mode



Figure 6: Pearson value compares (GOSAT XCH₄ / TROPOMI XCH₄) and surface albedo SWIR data. -0.1 < Pearson values < 0.1 are considered "no correlation" and are the ideal goal.



Month of Yea Figure 7: Agricultural land-use a) is reflected in albedo corrected CH₄ columns for summer c) and winter d). The magnitude of correction, b), has a clear seasonal cycle and is dependent on crop type.

- with other top-down estimates.



Figure 8: Effects of seasonal albedo correction on CH4 emission retrieval from the Denver-Julesburg basin.

 \rightarrow CH₄ columns contain a significant seasonal albedo bias that is successfully addressed with our new, seasonal bias correction.





Figure 4 (left) and 5 (top): Results of sensitivity study on deriving methane emissions from 2018-2022 CH₄ VCDs over the Permian basin, separated by boundary layer wind

 \rightarrow The long lifetime of methane (~9 years) creates biases in emissions derived with the divergence technique when the boundary layer is not efficiently vented.

Analysis of albedo corrected methane columns finds:

• Albedo corrected methane data shows dependence on underlying land-use structures, where different kinds of agriculture (water intensive crops: corn, sugarbeets, etc. drought resistant crops: winter wheat, dry beans, etc) tend to require different correction values (Fig 7a/c/d).

• Average correction values over each land-use type are distinctly different and show seasonality across the entire year, with larger corrections needed in the summer and smaller corrections needed in the winter (Fig. 7b).

Effect of seasonal albedo correction on derived emissions is:

• Preliminary! The Denver-Julesburg basin is a small area with comparatively low data statistics, which can lead to artefacts in retrieved emissions. Furthermore, winds and data statistics vary by season as well.

• Significant. Retrieved emissions are generally lower (Fig. 8) and more in line