

Author response to reviewer comments

Anonymous Reviewer #2

The authors present results from many MethaneAIR flights performed in the United States, primarily to quantify oil&gas emissions across major basins. Overall this represents a tremendous body of work to execute the flights, process data, and analyze for fluxes and the authors should be commended for this effort. The manuscript succinctly summarizes the results and shows general consistency in emission rates derived from previous observation studies (mass-balance, and satellite remote sensing). The authors employ what appears to be a novel way to calculate methane fluxes from remote sensing observations, but the details are extremely light.

Response- We thank the reviewer for their helpful comments and edits, and hope the following responses address their concerns. Our point by point responses to all comments are below in blue, with the page numbers corresponding to the attached revised manuscript with tracked changes included.

Before I can accept for publication, considerable more detail needs to be included and justified. My comments are as follows:

1. Table 1. Is Basin Area the total area of the basin or total area flown? If total area, can you express in the same table how much of that area you flew with MethaneAIR?

Response- The basin area in Table 1 is indeed the total area, not the total area flown. We have removed this column in Table 1 as it is not used in our analysis. In Table 3 in the main text, we added the area (in km²) covered by the MethaneAIR flights in each oil and gas basin. We also report the fraction of the basin's total production covered during the MethaneAIR flights, which is a more informative metric than the percent of the basin's area flown, since some basins have significant geographic coverage but production is concentrated in smaller regions which is where we would expect the majority of methane emissions to originate from.

Section S1. STILT.

2. How do you simulate columns with STILT? How many layers? Interpolate between layers? Use an averaging kernel? What is the averaging kernel?

Response- We have expanded the Supplement (Section S1) to add details about the column integration of the STILT model. In short, we ran STILT with 300 particles at the center of every meteorological model layer for each of the GFS and HRRR model up to 3x the planetary boundary layer height with integration using the fraction of total atmosphere column dry air and the mean MethaneAIR averaging kernel (see Chan Miller et al., 2024) as the weighting function. Section S1 now includes the following details:

Section S1, p.3: *“The Jacobian was computed using the Stochastic Time-Inverted Lagrangian Transport (STILT) model (Fasoli et al., 2018; Lin et al., 2003), which simulates the sensitivity of XCH₄ observations to sources on the ground by propagating air parcel trajectories backwards in time. The Jacobian was computed on a 0.01° x 0.01°*

grid over a $10^\circ \times 10^\circ$ domain around the center of the flight with trajectories long enough to fully exit the domain or include the previous day's boundary layer (28 hours backtime). Where possible, the Jacobian was computed twice, 1) with STILT driven by meteorological data from the operational Global Forecast System (GFS) model and 2) with STILT driven by meteorology from the High-Resolution Rapid Refresh (HRRR) model. Meteorological data was provided by the NOAA ARL meteorological archives in ARL format (<https://www.ready.noaa.gov/archives.php>). STILT was run as a column receptor, with a receptor placed at every layer of the meteorological input from the surface to 3x the planetary boundary layer height (above which we assume the footprint is always 0). STILT footprints for every layer are integrated with weights representing the fraction of the total atmospheric column of dry air represented, with the mean averaging kernel for MethaneAIR."

Section S1. The calculation of the background is unclear.

Response- We have substantially expanded the Supplement (Section S1) to add details about the background concentration calculation. In short, the background model is a field of methane concentrations taken from the MethaneAIR L2 product (see Chan Miller et al., 2024), fitted from below to the observations, with an allowance for the instrument precision.

Section S1, p.3: "The background concentration field represents the synoptic-scale, topographically varying component of the XCH_4 observations. We fit a field of background XCH_4 concentrations given by the MethaneAIR L2 prior (Chan Miller et al., 2024) from below, such that the reflected distribution of concentrations below background have a variance that matches the instrument precision. The MethaneAIR L2 prior forms a surface that varies realistically with topography in accordance with the vertical distribution of methane in the atmosphere from GEOS-FP Reanalysis (Rienecker et al., 2008) and the high-resolution digital elevation map tiles from Amazon Web Services (Larrick et al., 2020). Emissions are reported in a truncated domain of interest within the concave hull of the observations."

3. Can you restate in terms of an equation, figure, or additional clarifying language? - "The background concentrations are given by a model..." - what model? STILT?

Response- We have expanded the Supplement (Section S1) to add details about the background concentration calculation. See above response for revised text added to Section S1.

4. "The boundary inflow is modeled using the Jacobian and emission rates outside the domain of observed concentrations." Where do you get these emissions? An inventory? Proper background quantification is so vital to robust inversions, this section needs to be much clearer.

Response- The boundary inflow is computed as pseudo-emissions outside the domain of interest but inside the total domain of the inversion. These are "pseudo-emissions" that represent actual emissions just outside the domain or mesoscale variability in the inflowing methane concentration. This section has been made clearer:

Section S1, p.3: "Boundary inflow "pseudo-emissions" are the component of the dispersed area source emissions inside the full $10^\circ \times 10^\circ$ domain but outside the domain of interest. We refer to them as "pseudo-emissions" since they represent any source of sub-synoptic scale variation in the inflowing methane field, whether from mesoscale background variation or inflow of sources just outside the domain of interest."

Section S1. Point Sources.

5. Is the divergence integral method to calculate point sources applied at the 0.01 binning or at the native resolution? If you are binning, then you are certainly subtracting out more than point sources, as you are aggregating all true emission sources within that ~1km domain.

Response- We are indeed binning. The divergence integral integrates emissions from an effective area of approximately 1 km² for MethaneAIR. The divergence integral integrates all emissions in this area, and so fixing the emissions in this gridcell is the appropriate choice to best represent the computed discrete sources.

6. If you are not binning, how do you assess that model transport error correctly subtracts the influence of point sources? Do you have a quality control approach that ensures this? If the transport is wrong, then you risk not subtracting the point source component in your concentration field, which I can imagine will throw the inversion haywire and produce flux artifacts.

Response- See above response regarding binning. If the transport is substantially wrong such that the total emissions in a flight are substantially affected, the flight fails quantity control and is excluded from the analysis.

7. How do you determine an origin of the point source via the divergence integral method? It seems quite critical that you get the origin correct if you are forward simulating a concentration field with STILT.

Response- The plume detection method first calculates the flux divergence for 600 m x 600 m squares tiled across the scene, using HRRR wind fields and the divergence integral method to calculate the flux through each square. The upwind end of each plume shows a higher flux divergence than the downwind end; hotspots in the gridded flux product were identified with a thresholding method as potential plume origins. At each flux hotspot, we found XCH₄ clumps with a given number of contiguous pixels above a threshold value to create a mask of the plume. We calculated the major axis of the XCH₄ mask and took the upwind end of the major axis (using the HRRR wind direction) to be the plume origin. All plumes found using this method were reviewed manually by examining plume morphology, ground infrastructure, and correlation with albedo, and false positives were discarded.

We have modified the text in the Supplement (Section S1) to include more detail on the method, as follows:

Section S1, p.2: *“For each MethaneAIR flight, discrete point source emissions (with methane emission rates > ~200 kg/hr), are detected using an automated threshold-based method with manual QA/QC prior to their inclusion in our analysis and subsequently quantified using a divergence integral (DI) method (Chulakadabba et al., 2023; Warren et al., 2025). The plume detection method first calculates the flux divergence for 600 m x 600 m squares tiled across the scene, using High-Resolution Rapid Refresh (HRRR) wind fields and the divergence integral method (Chulakadabba et al., 2023) to calculate the flux through each square. In the gridded flux product, hotspots were identified with a thresholding method as potential plume origins. At each flux hotspot, we found XCH₄ clumps with a given number of contiguous pixels above a threshold value to create a mask of the plume. We calculated the major axis of the XCH₄ mask and took the upwind end of the major axis (using the HRRR wind direction) to be the plume origin (Warren et al., 2025). This system has been validated with controlled release experiments (Chulakadabba et*

al., 2023; El Abbadi et al., 2024), and is explained in greater detail in Chulakadabba et al., 2023 and Warren et al., 2025.”

8. It's not obvious to the reader that subtracting a forward model simulated concentration field produces a preferred result, especially assuming some level of spatial aggregation (e.g., were you to run these inversions at 10-20km, scales that others perform satellite inversions, would this still be required?).

Response- Rather than subtracting a forward model simulated concentration field we fix the emissions in the gridcell where the discrete source was detected. This is the most accurate interpretation of the quantity measured by the divergence integral algorithm.

Section S1. Inverse Problem

9. What is the formulation of the inversion problem? An SI is a good place to put these equations to paper. The way it currently reads, the mass-balance constraint would just be the inverse of the Jacobian - i.e., $s = (H^{-1})y$ and the non-negativity constraint would be some sort of gradient descent (or something else?) algorithm that stops at zero. Not obvious from what's written. Are there other parameters that keep the solution from an overfit? They say there is no need for a prior, so not Bayesian I guess?

Response- We have substantially increased the amount of detail (revised text below) about the inverse model in the Supplement (Section S1).

Section S1, p. 2-4: “With discrete sources (s_{discrete}) fixed, the inverse model fits a gridded field of dispersed area source emission rates ($s_{\text{dispersed}}$) to account for the balance of the methane enhancement. A gridded field of emission rates in the domain of interest, ($s_{\text{reported}} = s_{\text{discrete}} + s_{\text{dispersed}}$), and “pseudo-emission” rates in the upwind boundary inflow region (s_{inflow}) are fitted to observed column-averaged dry-air mole fractions of methane (XCH_4), z , linked by a Jacobian (H) plus a field of background concentrations (b) (Equation S1).

$$z = H(s_{\text{discrete}} + s_{\text{dispersed}} + s_{\text{inflow}}) + b \quad (\text{Eq. S1})$$

The inversion enforces non-negative fluxes and exact conservation of the observed methane mass to maintain physical realism and applies Tikhonov regularization to promote spatial smoothness and mitigate the sensitivity of the hybrid framework to transport errors and measurement noise. We solve for the non-negative emission field ($s = s_{\text{discrete}} + s_{\text{dispersed}} + s_{\text{inflow}}$) that reproduces the MethaneAIR enhancements:

$$J_s = \|H_s - z - b\|^2 + \lambda^2 \|L(s)\|^2 \quad (\text{Eq. S2})$$

st. $s \geq 0, \quad w^T(H_s) = M,$

where:

L – first-order spatial difference operator enforcing smoothness

λ – Tikhonov regularization strength

M – total methane mass enhancement in the domain (kg CH_4)

w – air-mass weights converting ppb to methane mass

XCH₄ observations were aggregated to 0.01° x 0.01° while preserving their location in time (allowing for overlapping observations from successive flight tracks). Aggregated grid cells at least 50% covered with data that passed all QA/QC flags were included in the analysis.

The Jacobian was computed using the Stochastic Time-Inverted Lagrangian Transport (STILT) model (Fasoli et al., 2018; Lin et al., 2003), which simulates the sensitivity of XCH₄ observations to sources on the ground by propagating air parcel trajectories backwards in time. The Jacobian was computed on a 0.01° x 0.01° grid over a 10° x 10° domain around the center of the flight with trajectories long enough to fully exit the domain or include the previous day's boundary layer (28 hours backtime). Where possible, the Jacobian was computed twice, 1) with STILT driven by meteorological data from the operational Global Forecast System (GFS) model and 2) with STILT driven by meteorology from the High-Resolution Rapid Refresh (HRRR) model. Meteorological data was provided by the NOAA ARL meteorological archives in ARL format (<https://www.ready.noaa.gov/archives.php>). STILT was run as a column receptor, with a receptor placed at every layer of the meteorological input from the surface to 3x the planetary boundary layer height (above which we assume the footprint is always 0). STILT footprints for every layer are integrated with weights representing the fraction of the total atmospheric column of dry air represented, with the mean averaging kernel for MethaneAIR.

The background concentration field represents the synoptic-scale, topographically varying component of the XCH₄ observations. We fit a field of background XCH₄ concentrations given by the MethaneAIR L2 prior (Chan Miller et al., 2024) from below, such that the reflected distribution of concentrations below background have a variance that matches the instrument precision. The MethaneAIR L2 prior forms a surface that varies realistically with topography in accordance with the vertical distribution of methane in the atmosphere from GEOS-FP Reanalysis (Rienecker et al., 2008) and the high-resolution digital elevation map tiles from Amazon Web Services (Larrick et al., 2020). Emissions are reported in a truncated domain of interest within the concave hull of the observations.

Boundary inflow “pseudo-emissions” are the component of the dispersed area source emissions inside the full 10° x 10° domain but outside the domain of interest. We refer to them as “pseudo-emissions” since they represent any source of sub-synoptic scale variation in the inflowing methane field, whether from mesoscale background variation or inflow of sources just outside the domain of interest.

Discrete sources are fixed in the area source inversion, fixing emissions in a 0.01° x 0.01° area, which approximates the effective representative area of the DI. This places trust in the well-tested point-source specific algorithm to do the best job at quantifying point source emissions and uses the Jacobian to ensure the complete mass of methane from the point sources are accounted for. The alternative method of plume-masking is inconsistent between methodologies and inevitably undercounts the contribution of the point sources when they fall below detectable concentrations. The inclusion of fixed discrete sources in the inverse model makes this a constrained regularized optimization rather than Bayesian inference, and so we report only the optimal solution and do not assign probabilistic confidence intervals. It is important to note that this is non-Bayesian to prevent over-interpretation of a “posterior” estimate, which would be invalid due to data re-use.

The inverse problem is then solved numerically using projected, limited memory, bounded Broyden-Fletcher-Goldfarb-Shanno (L-BFGS-B) algorithm. The solution is initialized to a flat field that satisfies the mass constraint. Subsequent proposals are constrained to be non-negative and satisfy the mass constraint.”

10. The authors do not provide any sort of metric of goodness of fit (e.g., $H * \hat{s}$ plotted against y) or information content from the retrieval. It's fairly common practice to show how well your model around the optimal emission state compares to observations. It's also common to show information content

metrics (e.g., degrees of freedom for signal, model-resolution matrices, etc) for inversions, but given there's not an explicit inverse formulation in paper, it's hard to know if that would be feasible.

Response- We have added significant details about the inversion methodology in the Supplement Section S1. As included in our further details in Section S1 and in our responses, this analysis is based on a constrained regularized optimization rather than Bayesian inference. We have also added a plot (Figure S5) showing the observed vs modeled XCH₄ enhancement for MethaneAIR flights.

11. Table S5. Please provide citations for literature based estimates, perhaps as an additional column in the table or a footnote.

We added the citations for the literature-based estimates in Table S5.

References:

Chan Miller, C., Roche, S., Wilzewski, J. S., Liu, X., Chance, K., Sour, A. H., Conway, E., Luo, B., Samra, J., Hawthorne, J., Sun, K., Staebell, C., Chulakadabba, A., Sargent, M., Benmergui, J. S., Franklin, J. E., Daube, B. C., Li, Y., Laughner, J. L., Baier, B. C., Gautam, R., Omara, M., and Wofsy, S. C.: Methane retrieval from MethaneAIR using the CO₂ proxy approach: a demonstration for the upcoming MethaneSAT mission, *Atmospheric Measurement Techniques*, 17, 5429–5454, <https://doi.org/10.5194/amt-17-5429-2024>, 2024.

Chulakadabba, A., Sargent, M., Lauvaux, T., Benmergui, J. S., Franklin, J. E., Chan Miller, C., Wilzewski, J. S., Roche, S., Conway, E., Sour, A. H., Sun, K., Luo, B., Hawthorne, J., Samra, J., Daube, B. C., Liu, X., Chance, K., Li, Y., Gautam, R., Omara, M., Rutherford, J. S., Sherwin, E. D., Brandt, A., and Wofsy, S. C.: Methane point source quantification using MethaneAIR: a new airborne imaging spectrometer, *Atmospheric Measurement Techniques*, 16, 5771–5785, <https://doi.org/10.5194/amt-16-5771-2023>, 2023.

El Abbadi, S. H., Chen, Z., Burdeau, P. M., Rutherford, J. S., Chen, Y., Zhang, Z., Sherwin, E. D., and Brandt, A. R.: Technological Maturity of Aircraft-Based Methane Sensing for Greenhouse Gas Mitigation, *Environ. Sci. Technol.*, 58, 9591–9600, <https://doi.org/10.1021/acs.est.4c02439>, 2024.

Fasoli, B., Lin, J. C., Bowling, D. R., Mitchell, L., and Mendoza, D.: Simulating atmospheric tracer concentrations for spatially distributed receptors: updates to the Stochastic Time-Inverted Lagrangian Transport model's R interface (STILT-R version 2), *Geoscientific Model Development*, 11, 2813–2824, <https://doi.org/10.5194/gmd-11-2813-2018>, 2018.

Larrick, G., Tian, Y., Rogers, U., Acosta, H., and Shen, F.: Interactive Visualization of 3D Terrain Data Stored in the Cloud, in: 2020 11th IEEE Annual Ubiquitous Computing, Electronics & Mobile Communication Conference (UEMCON), 2020 11th IEEE Annual Ubiquitous Computing, Electronics & Mobile Communication Conference (UEMCON), 0063–0070, <https://doi.org/10.1109/UEMCON51285.2020.9298063>, 2020.

Lin, J. C., Gerbig, C., Wofsy, S. C., Andrews, A. E., Daube, B. C., Davis, K. J., and Grainger, C. A.: A near-field tool for simulating the upstream influence of atmospheric observations: The Stochastic Time-Inverted Lagrangian Transport (STILT) model, *Journal of Geophysical Research: Atmospheres*, 108, <https://doi.org/10.1029/2002JD003161>, 2003.

Rienecker, M. M., Suarez, M. J., Todling, R., Bacmeister, J., Takacs, L., Liu, H.-C., Gu, W., Sienkiewicz, M., Koster, R. D., Gelaro, R., Stajner, I., and Nielsen, J. E.: The GEOS-5 Data Assimilation System-Documentation of Versions 5.0.1, 5.1.0, and 5.2.0, 2008.

Warren, J. D., Sargent, M., Williams, J. P., Omara, M., Miller, C. C., Roche, S., MacKay, K., Manninen, E., Chulakadabba, A., Himmelberger, A., Benmergui, J., Zhang, Z., Guanter, L., Wofsy, S., and Gautam, R.: Sectoral contributions of high-emitting methane point sources from major US onshore oil and gas producing basins using airborne measurements from MethaneAIR, *Atmospheric Chemistry and Physics*, 25, 10661–10675, <https://doi.org/10.5194/acp-25-10661-2025>, 2025.