Author response to reviewer comments

Anonymous Reviewer #1

The authors quantify methane emissions from 12 US oil and gas basins using methane column observations from 32 MethaneAIR flights in 2023. These 12 basins accounted for 70% of total onshore oil and gas production in the contiguous United States in 2023. The authors estimate both total and sector-specific (oil + gas) emissions for each basin. They use a novel two-step regional flux inversion approach that first quantifies large point sources and then diffuse area emissions via Bayesian inverse analysis with the Stochastic Time-Inverted Lagrangian Transport (STILT) model. Emission contributions from non-oil and gas sources are estimated using sectoral emission estimates from a collection of previous top-down and bottom-up studies. The authors compare their regional estimates of methane emissions and loss rates with 16 previous studies and find generally good agreement.

We thank the reviewer for their helpful comments and suggestions. We have addressed the reviewer comments and incorporated them in the revised manuscript 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 revised manuscript with tracked changes included.

The manuscript is well-written and a good fit for ACP. I recommend that it be accepted for publication with revisions to address the following comments and questions:

I think a more detailed description of the flux inversion methodology is needed, ideally in the main text. It is a novel approach and the most critical part of the analysis. Can more information be provided? For example, it would be helpful to know more about the modeling of the "boundary inflow", the numerical solution to the inverse problem, the approach to calculating column sensitivities with STILT (presumably based on MethaneAIR retrieval averaging kernels), and how/why GFS and HRRR meteorology are combined to drive STILT.

Response-We have substantially expanded Section S1 in the Supplement to describe the inversion framework in detail. We added methodological details and equations for the forward model, the background, the boundary inflow, and the solution to the inverse problem.

We clarified that independent Jacobians were computed using 1) GFS meteorology, and 2) HRRR meteorology where possible. These were used to diagnose potential issues with excessive transport error (diverging cases were excluded by QA/QC), and in the estimation of uncertainty. The following text is now included in 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 ($s_{dispersed}$) to account for the balance of the methane enhancement. A gridded field of emission rates in the domain of interest, ($s_{reported} = s_{discrete} + s_{dispersed}$), 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).*

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

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 ($s = s_{discrete} + s_{dispersed} + s_{inflow}$) *that reproduces the MethaneAIR enhancements:*

$$J_s = \|H_s - z - b\|^2 + \lambda^2 \|L(s)\|^2 \quad \text{(Eq. S2)}$$
$$\text{st.} \quad s \geq 0, \quad w^T(H_s) = M,$$

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

XCH_4 *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 XCH_4 *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 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.*

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."

What happens to the flux inversion if the modeled wind direction for a point source is wrong? In principle a flux dipole could arise, but it is mentioned in the SI that the non-negativity constraint in the inversion helps prevent that. How well does that work, and do any other biases manifest?

Response-When the modeled wind direction is wrong, plumes downwind of point sources are poorly modeled and a "plume shadow" or "dipole" effect is induced (a dipole is technically the result if non-negativity is not enforced). In cases where the modeled plume does not overlap the observed plume, the Jacobian is excluded by QA/QC. If both GFS and HRRR Jacobians are excluded by QA/QC, then the flight is excluded by QA/QC. There will always be some transport error, which is exacerbated where there are steep gradients in methane enhancements, most notably where there are distinct plumes. This is not only because of error in the mean wind direction, but also because of variations in XCH4 that cannot be resolved by the meteorological model (i.e., large eddies).

The inverse model uses a hard mass constraint Tikhonov regularization (more information about this has been added to the Supplement Section S1 – see above response for added text). This hard mass constraint ensures that the total excess methane in the observed atmosphere is modelled, and the spatial allocation of the inverse model distributes area emissions spatially throughout the region. The impact of wind error in the vicinity of large point sources is then to re-distribute other emissions sources around the plume, with minimal effect on the total, as the residence time of air is mostly unchanged by this perturbation.

How are point sources detected prior to the diffuse flux inversion? Is the process automated, semi-automated, or manual?

Response- Point sources are detected using an automated threshold-based method, with manual QA/QC prior to their inclusion in our analysis. The process is described in detail in Chulakadabba et al. 2023 and Warren et al. 2025 and validated in controlled release experiments (El Abbadi et al. 2024; Chulakadabba et al. 2023). We have expanded Section S1 in the Supplement 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 XCH_4 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 XCH_4 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 Warren et al., 2025."*

305-310: There is some redundant content in this passage.

Response- This passage importantly describes both the total methane emissions estimated by MethaneAIR as well as the oil and gas only methane emissions estimated by MethaneAIR, and how these two different estimates compare to the EPA totals. While the discrepancy between the overall total and oil and gas total is similar, we believe including both estimates and comparisons is valuable, as it shows that the oil and gas sector is likely the main contributor to underreported emissions in the GHGI.

314: 0.17 kg CH4/GJ from MethaneAIR is very similar to 0.18 kg CH4/GJ from IEA. Is it expected to be much lower? Perhaps this passage can be clarified.

Response- We modified the text to add clarity to the comparison between the MethaneAIR and IEA intensity estimates, as there are other important differences between the two in addition to the use of gross vs. marketed gas production that should be mentioned. Considering these other factors, we do not expect the IEA value to necessarily be lower than our MethaneAIR estimate, so we modified the text as follows:

L312-315: *"The estimated energy-normalized methane intensity of 0.17 kg CH_4/GJ is comparable to the upstream methane intensity of 0.18 kg CH_4/GJ for the entire US reported by the IEA for 2024 (IEA, 2025), however it should be noted that their estimate is calculated using marketed oil and gas production, whereas our estimate uses gross production and includes methane emissions from the entire oil and gas sector (i.e., not just upstream)."*

319: I believe Figure 6 is mislabeled here—the passage seems to refer to Figure 5.

Response- We double checked the caption for Figure 6 and confirmed that there is no labelling error, it describes the comparison of MethaneAIR derived loss rates to other measurement-based loss rates from previous literature. Note that there are some similarities in the features for Figures 5 and 6 (e.g., the grey shaded area and dashed lines), hence the similar descriptions in the captions.

Figures 5 and 6:
In which cases are the MethaneAIR and previous estimates for the same domain? Is the Zhang et al. result for the Permian spatially resampled to the flight domain? Those authors reported a loss rate of 3.7%, much higher than the <2% shown in Figure 6, so I assume so. It would be helpful to mark on the plots whether or not the previous results reflect spatial resampling.

Response- We added an asterisk to the x-axis labels and expanded the caption text to note which previous estimates correspond to the exact domain, and which ones correspond to similar/overlapping areas. Regarding the Zhang et al., 2020 reported loss rate, the difference is related to the domains - their reported loss rate reflects their entire study domain (i.e., the whole Permian basin) whereas the loss rate we show in the figure was computed based on Zhang et al.'s reported emissions and production volumes within the MethaneAIR flight domain which is a subset of the Permian basin.

Why are the x labels in the figures not identical? There are fewer bars in some subplots of Figure 6 than Figure 5.

Response- There are a different number of bars in the figures because some studies only reported total methane emissions and did not report methane loss rates or the necessary information (e.g., gas production volumes at the time of measurement) for us to compute it for our analysis. In these cases, they are included in Figure 5 but not included in Figure 6.

Why do the inter-study differences in methane emissions not more closely match the differences in methane loss rate? Two examples of this: the Peischl bars in the Barnett subplots show much higher emissions than MethaneAIR but very similar loss rate, and the MethaneAIR bars in the Permian subplots show better agreement with previous studies for emissions than loss rate.

Response- Absolute emissions can have more variability due to changes in activity/production levels over time, whereas methane loss rates are often more stable over time as they normalize emissions by production. This is likely contributing to the observed difference in the Peischl et al., 2018 study in the Barnett, as those measurements were collected in 2015 when oil and gas production was much higher than when the MethaneAIR flights took place in 2023. For the Permian, the time of measurement for many of the studies is similar, likely contributing to the better agreement in total emissions. We also argue that the loss rates show similarly good agreement for the Permian (<1% difference across all studies).

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