**High-resolution North American methane emissions inferred from an inversion of 2019 TROPOMI satellite data**

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We quantify 2019 North American methane emissions at 0.25° ⨉ 0.3125° resolution by inverse analysis of atmospheric methane columns measured by the Tropospheric Monitoring Instrument (TROPOMI). We perform substantial cleaning of the TROPOMI data to avoid regional biases associated with snow and ice cover. Prior methane emission estimates for the inversion are given by gridded versions of the national inventories reported by individual countries under the Paris Agreement in 2015 (Mexico), 2012 (the United States), and 2018 (Canada). We update United States oil and natural gas emission to match 2018 infrastructure and emissions. We generate an ensemble of improved emissions estimates through analytical minimization of a Bayesian cost function, yielding explicit quantification of the information content of the observing system. We achieve high resolution results with a reduced-rank eigenvector characterization of the observing system that maximizes information content while decreasing computational cost by an order of magnitude. Our reduced-rank inversion optimizes 65% of prior emissions estimates across North America and 75% of prior emissions in the contiguous United States. We report the ensemble spread for sectoral, state, and urban emissions. We find an underestimate of livestock emissions that correlates with the distribution of hog farms, suggesting underestimated manure management emissions. We also find an 20-25% underestimate in urban emissions that can be attributed either to underreported landfill emissions or post-meter natural gas emissions. The significant increase in resolution achieved while quantifying the inversion information content demonstrates the potential to improve the attribution of inferred greenhouse gas emissions to sectors, nations, states, and urban areas.

**1 Introduction**

All modeled pathways that prevent global warming above 1.5°C require methane emissions reductions (IPCC SR5). The United Nations Framework Convention on Climate Change (UNFCCC) requires member parties to report methane emissions. The “bottom-up” approaches used to generate these inventories estimate emissions using information on sectoral activity levels and emission factors. However, considerable uncertainty exists in the spatial and temporal distribution of sectoral emissions and emission factors. Satellite, aircraft, and in situ observations can evaluate inventories through inverse analyses. To accurately quantify sectoral emissions at a national or subnational level, inversions must both achieve high resolution and quantify the information content of the observing system. Here we use column methane observations from the TROPOMI satellite instrument to infer methane emissions and the associated information content at 0.25° ⨉ 0.3125° resolution over North America for 2019, allowing detailed analysis of sectoral, state, and urban emissions.

Inverse analyses optimize methane emissions estimates by fitting observations to simulated concentrations from a chemical transport model (CTM). When a linear relationship exists between emissions and concentrations, the optimal (posterior) solution can be found by numerically or analytically minimizing a Bayesian cost function regularized by a prior emissions estimate. The numerical (variational) approach uses gradient descent algorithms together with the adjoint of the CTM. The computational cost of this approach scales less-than-exponentially with the resolution of the emissions estimate, but the computational cost can still be prohibitive at high resolution due to the time required for the solution to converge (U of M). Moreover, the ensemble or low-rank approaches used to estimate the posterior error and information content are computationally expensive and often incomplete. The analytical approach provides closed-form expressions for the posterior emissions and the associated error and information content. However, the computational cost scales more-than-exponentially with the resolution of the optimized emissions due to the cost of constructing the Jacobian matrix that characterizes the linear relationship between emissions and concentrations in the CTM.

Past inversions achieved high-resolution at the national level using variational approaches, dimension reduction techniques, or by solving the inversion at coarser resolution and using optimal regridding schemes.

* Adjoint
* Dimension reduction (subsetting domain or aggregating together grid cells)
* Cusworth approach
* Intro outline
* Don’t need to explain why we care about methane—look at IMI as an example
* Climate agreements—need to go to higher resolution to quantify country-level emissions, requiring continental scale
  + Issuue with sector attribution and geographic atttribution (maybe cite Tia’s paper)
* Next paragraph: how we’ve achieved high resolution in the past (adjoint, GMMs, Cusworth approach)

[define observing system]

Inverse studies summary paragraph

Papers to cite

NA inversions

* Miller et al. 2013 – Lagrangian inversion of observations from towers and aircraft, analyzes footprints for each of 12,694 observations, geostatistical inversion (no prior), 1deg 1deg
* Wecht et al. 2014 – SCIAMACHY, adjoint
* Turner et al. 2015 – GOSAT, GMM, EDGAR
* Janardanan et al. 2017?
* Bruhwiler et al. 2017?
* Sheng et al. 2018?
* Lan et al. 2019?
* Maasakkers et al. 2021 – GOSAT, GMM, EPA GHGI

High resolution regional inversions

* Wecht et al. 2014 – analytical inversion over western North America and Pacific? (157 grid cells at 0.5 degrees) (Spatially resolving methane emissions in California, ACP)
* Sheng et al. 2018 – Regional SEAC4RS using GMM
* Zhang et al. 2020 – Permian inversion using analytical inversion

Inversion at 25 km resolution enabled by reduced-rank approach

Paragraph on the reduced-rank approach

**2** **Data and methods**

We conduct an inversion of 2019 TROPOMI methane observations over North America. The TROPOMI observations are fit to simulated concentrations from the GEOS-Chem chemical transport model (CTM, [www.geos-chem.org](http://www.geos-chem.org)) to optimize mean methane emissions at 0.25° ⨉ 0.3125° spatial resolution. We calculate the optimal emissions and the associated error covariance and information content by finding the analytical minimum of a Bayesian cost function regularized by a prior emissions estimate (section 2.1). Sections 2.2 through 2.6 describe the components of the inversion: section 2.2 describes the state vector, prior emissions, and prior errors; section 2.3 describes GEOS-Chem; section 2.4 describes TROPOMI observations; section 2.5 describes the observing system errors; and section 2.6 describes the novel, reduced-rank method used to calculate the Jacobian matrix. Section 2.7 summarizes the inversion ensemble. Section 2.8 describes the method used to attribute the posterior emissions to sectors, states, and urban areas.

**2.1 Reduced-rank analytical inversion**

We optimize the state vector of gridded emissions assuming normal errors by minimizing a Bayesian cost function

where and are the prior emissions estimate and error covariance matrix, respectively (section 2.4); and are the vector of observations and the error covariance matrix, respectively (section 2.5); is the chemical transport model (CTM) that simulates observations as a function of emissions (section 2.2); and is a regularization factor that accounts for the absence of covariance in (section 2.4) (Brasseur and Jacob, 2017). The nested methane CTM is linear so that where is the Jacobian matrix and is constant. Analytical minimization of the cost function then yields the optimal (posterior) state vector estimate , error covariance matrix , and information content given by the averaging kernel matrix , which describes the sensitivity of the posterior estimate to the true state vector. The diagonal elements are commonly referred to as averaging kernel sensitivities and their sum gives the degrees of freedom for signal (DOFS), the number of pieces of information independently constrained by the observing system (Rodgers, 2000). We generate an inversion ensemble using a range of prior emissions, prior errors, observations, and observing system errors, giving an additional estimate of inversion error (section 2.7).

The standard analytical solution is numerically unstable for large since it requires inverting a non-sparse matrix. We use reduced-rank approximations that solve the inversion on an orthonormal basis that optimally spans the information content of the satellite–forward model observing system (Bousserez and Henze, 2018). The basis is given by the eigendecomposition of the prior-preconditioned Hessian

where the columns of are the eigenvectors and the diagonal of gives the eigenvalues. Then,

Here, is the full-rank approximation that minimizes error relative to the standard solution; and are the posterior error covariance matrix and averaging kernel matrix associated with the reduced-rank Jacobian matrix (section 2.6), respectively; is the matrix of the first eigenvectors; and is a diagonal matrix containing the largest eigenvalues. We choose = 1952 to match the rank of the Jacobian matrix. To reduce errors associated with the rank reduction, we filter out grid cells with averaging kernel sensitivities less than 0.05. The calculation of the prior pre-conditioned Hessian still requires significant memory for large and . We parallelize the calculation manually and using Dask, a python program that performs parallelization for datasets that exceed available memory.

**2.2 State vector, prior estimate, and prior error**

In all inversions, we optimize emissions in 23,691 grid cells at 0.25° ⨉ 0.3125° resolution over North America, including all grid cells containing land or anthropogenic methane emissions larger than 0.1 Mg km-2 a-1, representing over 99% of methane emissions in North America. We also conduct inversions that optimize boundary four cardinal (north, south, east, and west) boundary condition elements. Methane chemical and soil sinks are not optimized because these loss processes are slow compared to the ventilation timescale.

Figure 1 shows the spatial distribution of major source sectors in the prior emissions estimate. Anthropogenic emissions for the United States, Mexico, and Canada are given by the spatially disaggregated (gridded) versions of the EPA GHGI for 2012 (Maasakkers et al., 2016), the INECC inventory for 2015 (Scarpelli et al., 2020), and the ECCC estimates for 2018 (Scarpelli et al., 2021), respectively. To account for changes in the distribution and magnitude of oil and natural gas emissions in the United States since 2012, we update the distribution of production fields using 2018 DrillingInfo data and scale the total natural gas production, transmission, processing, and distribution emissions to match 2018 emissions as reported in the 2020 GHGI. We also use the Environmental Defense Fund’s high-resolution inventory over the Permian basin for xxxx, one of the largest oil and natural gas producing regions in North America (Zhang et al., 2020). Because this inventory does not differentiate between oil and natural gas emissions due to the challenges of separating the sources, we treat oil and natural gas as a single sector in our analysis. All other anthropogenic emissions in the North American domain are provided by the EDGAR v4.3.2 global emission inventory for 2012 (?). Anthropogenic emissions are assumed aseasonal except for manure management and rice cultivation, for which we apply seasonal scaling factors as described by Maasakkers et al. (2016) and Zhang et al. (2016), respectively.

Wetlands are the dominant natural source of methane emissions, but large uncertainties exist in the distribution and magnitude of wetland emissions (Bloom et al., xxxx). We address this uncertainty by including two wetland inventories in our inversion ensemble. Figure 1 (bottom right) shows the scaled and subsetted versions of the high performance WetCHARTs ensemble version 1.3.1 used (Ma et al. 2021). Previous inversions found a large overestimate of wetland methane emissions in the high-performance ensemble, particularly in the boreal wetlands (Lu et al., 2022). The scaled inventory decreases total wetland emissions by a factor of 4.04 based on a comparison of the ensemble to FLUXNET CH4, a network of eddy covariance tower data (Shuang et al., ????). The subsetted inventory removes two ensemble members that produce anomalously large emissions in the high northern latitudes in summer and fall (Lu et al. 202?). Other natural methane emission sources include open fires, termites, and geological seeps. We use the same emissions for these sources as in Lu et al. (2022).

We assume uniform relative prior error standard deviations for emissions of between 50% and 100% for ach ensemble member. The 50% relative error lower bound follows previous inversions that optimized methane emissions over North America (Maasakkers et al., 2021; Lu et al., 2022). We increase errors up to 100% to account for displacement errors and increased error covariance at high resolution (Maasakkers et al. 2016). Ensemble members that optimize the boundary conditions use a base error standard deviation of 10 ppb, which is scaled by between 0.5 and 1 depending on the choice of grid cell prior error. We choose the prior error for each ensemble member as described in section 2.7. Although lognormal errors better represent the distribution of methane emissions and avoid negative posterior emissions (e.g., Maasakkers et al., 20?, Chen et al., 2022), the corresponding solution iteratively updates the Jacobian matrix, which is computationally prohibitive for our large observation and state vector dimensions (section 2.1). We instead use normal errors. In the absence of better information, we assume there is no error covariance.

**2.3 Forward model**

We use the nested version of the GEOS-Chem chemical transport model (CTM) v12.7 at 0.25° ⨉ 0.3125° resolution over North America as the forward model for the inversion. Earlier versions of the methane simulation were described by Wecht et al. (2014) and Turner et al. (2015). The model is driven by GEOS-FP meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO). Methane loss from OH, Cl, soil uptake, and stratospheric oxidation is described in Maasakkers et al. (2019). Initial conditions for January 2019 and 3-hourly boundary conditions for the year are given by the methane concentration fields from the global 2° ⨉ 2.5° TROPOMI inversion conducted by Qu et al. (2021). These concentration fields are unbiased with respect to the global TROPOMI data and are informed predominantly by observations outside of North America.

We validate GEOS-Chem by comparison to surface and aircraft methane observations for May 2018. We use observations from the Atmospheric Tomography Mission (ATom), the Atmospheric Carbon and Transport – America (ACT-America) campaign, and the NOAA Observation Package (ObsPack). We find a mean model-observation bias of 6.36 ppb and a correlation of R = 0.45. We also find no significant latitudinal bias in the model-observation difference, although the surface and aircraft observations provide significant coverage only between 30°N and 50°N.

**2.4 TROPOMI observations**

The Tropospheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5 Precursor satellite has provided daily, global observations of dry column methane mixing ratios at 7 ⨉ 7 km2 nadir pixel resolution since May 2018 and at 5.5 ⨉ 7 km2 nadir pixel resolution since August 2019 (citation). TROPOMI measures backscattered solar radiation at 2.3 μm from a sun-synchronous orbit with a local overpass time of 13:30 (Veefkind et al. 2012). TROPOMI retrieves methane concentrations using a full-physics retrieval, which is limited by cloud cover, variable topography, albedo, and high aerosol loading (citation). As a result, TROPOMI has a xx% retrieval rate over North America for 2019. We use the retrieval described by Lorente et al. (2021), which has a -3.4 ± 5.6 ppb bias relative to the Total Carbon Column Observing Network (TCCON). We use only high-quality retrievals as indicated by the quality assessment flag.

We evaluate the TROPOMI data using methane observations from the Greenhouse gases Observing SATellite (GOSAT). Launched in 2009, GOSAT provides high-precision observations of methane in 10 km diameter nadir pixels separated by ~250 km along- and cross-track. GOSAT measures backscattered solar radiation at 1.6 μm with a local overpass time of about 13:00 and a three-day return time. We use the GOSAT methane retrieval version 9.0 of the University of Leicester obtained by the CO2 proxy method (Parker and Boesch, 2020, last accessed 29 December 2020). We use only high-quality retrievals as indicated by the quality assessment flag. Due to the sparse coverage of GOSAT, we also evaluate the TROPOMI data using a GEOS-Chem simulation run with the prior emissions.

We compare average seasonal TROPOMI and GOSAT methane observations on a 2° ⨉ 2° grid following Lorente et al. (2021). We find large regional biases, defined as the standard deviation of the mean TROPOMI – GOSAT difference, of between 15 ppb (summer) and 20 ppb (winter). The winter-time biases are likely due to snow- and ice-covered scenes (Lorente et al. 2021). We identify these scenes using blended albedo, an empirical parameter that combines shortwave and near-infrared albedo and that correlates with snow- and ice-cover when greater than about 1 (Wunch et al. 2011). We remove scenes with blended albedo > 0.75 in fall, winter, and spring. We also remove scenes with shortwave albedo less than 0.05 following de Gouw et al., 2020. These scenes exhibit large prior GEOS-Chem – TROPOMI biases and disproportionately account for the remaining unphysical TROPOMI observations (XCH4 < 1700 ppb). Finally, we remove scenes north of 50°N in winter, which are likely to correspond with snow- and ice-cover and which exhibit anomalous prior GEOS-Chem – TROPOMI differences.

The applied filters increase the GOSAT-TROPOMI correlation in all seasons, with the largest increases in winter and spring (from R2 = 0.20 to 0.30 and from R2 = 0.32 to R2 = 0.49, respectively), when snow- and ice-cover are most likely to impact the TROPOMI retrieval. Seasonal regional biases decrease by between 7% and 21% and are in all cases less than the standard deviation of both the TROPOMI and GOSAT data. The mean TROPOMI – GOSAT biases are also consistent with the -10.3 ± 16.8 ppb bias found by Lorente et al. (2021).

After filtering the data, we find a residual aseasonal latitudinal bias in the TROPOMI – GEOS-Chem difference. This bias has been noted and corrected previously by Turner et al. (xxxx), Maasakkers et al. (xxxx), and Zhang et al. (xxxx). We define a latitudinal correction term (ppb) for the GEOS-Chem – TROPOMI difference using a first-order polynomial. For the inversion ensemble members that use scaled wetland emissions, we find , where is the degrees latitude. For the members that use the subsetted wetland emissions, we find . We also conduct sensitivity tests without the latitudinal correction. In these cases, we remove a mean bias of 8.53 ppb (scaled wetland emissions) and 9.11 ppb (subsetted wetland ensemble), which we attribute to errors in the boundary conditions.

Figure 2 (top row) shows the 2919358 final observations, regridded onto the GEOS-Chem grid and averaged seasonally, that constitute our observation vector **y**. The bottom row shows the observational density. We preserve 69% of the original high-quality data and find good agreement with the prior GEOS-Chem simulation (R between 0.58 and 0.60, depending on the wetland emissions prior and bias correction). We still find large, seasonally-variable gradients in the prior GEOS-Chem – TROPOMI difference (e.g., in spring over Northern Wisconsin), suggesting the possibility of residual systematic biases in the observations. We account for these biases in our observing system errors.

**2.5 Observing system errors**

Observing system errors include contributions from the forward model, the instrument, and representation error (Brasseur and Jacob, 2017). We calculate the variances using the residual error method (Heald et al. 2004). This method assumes that the mean difference between the TROPOMI observations and the prior GEOS-Chem simulation in each grid cell is caused by errors in emissions that will be corrected by the inversion. The standard deviation of the residual errors after subtracting the mean gridded errors then gives the observational errors. We calculate the seasonal mean difference on a 2° ⨉ 2° grid to minimize the effect of systematic albedo biases in the TROPOMI data. In the <100 scenes where the residual standard deviation is less than the reported instrument error standard deviation (Lorente et al., 2021), we use the latter instead. We also set a minimum error of 10 ppb, which applies to between 31 and 32% observations, depending on the prior wetland emissions and bias correction. We find a mean observational error standard deviation of 11.5 ppb in all cases, with larger errors in winter and at high latitudes. The resulting variances are the diagonal elements of our observational error covariance matrix . Off-diagonal terms are assumed zero in the absence of better information. We introduce a regularization factor , described in section 2.7, to account for the lack of covariance (Chevallier et al., 2007).

**2.6 Jacobian matrix**

The relationship between simulated methane concentrations and emissions in the nested version of GEOS-Chem is strictly linear and is described by the Jacobian matrix . The Jacobian matrix is typically constructed by conducting a forward model simulation for each state vector element. While this is an embarrassingly parallel problem, constructing this matrix for the 23691 0.25° x 0.3125° resolution grid cells optimized by this inversion is computationally intractable. We take advantage of the heterogeneous information content of the TROPOMI observations to construct the Jacobian matrix at substantially decreased computational cost using the reduced-rank method introduced by Nesser et al. (2021). This method updates an initial, low-cost estimate of the Jacobian matrix by perturbing the patterns that best explain the information content of the observing system rather than grid cells, constructing a reduced-rank Jacobian matrix while optimally preserving information content.

We construct the initial estimate of the Jacobian matrix using the mass-balance approach introduced by Nesser et al. (2021). We assume that a perturbation of methane emissions in grid cell *j* produces column mixing ratio enhancements in nearby observation *i* according to

where is a dimensionless, mass-conserving coefficient providing a crude representation of turbulent diffusion that decreases the sparsity of , and are the molecular weights of dry air and methane, respectively, is a ventilation length scale equal to the square root of the grid cell area, is gravitational acceleration, is the local wind speed taken here as 5 km h-1, and is the surface pressure taken here as 1000 hPa. We assume decreases exponentially as = {10, 6, 4, 3, 2.5} from the inner to the outer ring of grid cells surrounding *j*, normalized and divided by the number of grid cells in each ring.

We use to calculate the patterns of information content perturbed in the forward model. The corresponding averaging kernel matrix captures the dominant patterns of information content because of its the dependence on the prior error covariance matrix and on the observational density as quantified by the observational error covariance matrix and by the sparsity structure of (Nesser et al., 2021). The initial patterns of information content are then given by the eigenvectors of the averaging kernel matrix calculated as where is the th eigenvector of the prior-preconditioned Hessian (Bousserez and Henze, 2018). We perturb the = 434 eigenvectors that span 50% of the initial information content in the forward model. We apply an optimal operator that restores the original state dimension and minimizes information content loss to the resulting matrix to yield an updated reduced-rank Jacobian matrix estimate . We then recompute the eigenvectors, perturb the = 1952 eigenvectors that explain 80% of the information content, and construct the updated reduced-rank Jacobian matrix . This update scheme optimizes the information content of the posterior solution while reducing the computational cost by an order of magnitude (Nesser et al., 2021).

**2.7 Inversion ensemble**

The posterior error covariance matrix yielded by an analytical inversion underestimates errors by failing to account for errors in the inversion parameters (Yu et al., 2021). We estimate these errors and reduce the noise associated with the reduced-rank Jacobian approximation by generating a quality-controlled ensemble of inversions, summarized in table 1. We generate eight base inversions by varying (1) the choice of prior wetland emissions estimate (either the scaled wetland emissions or the modified wetland ensemble), (2) the inclusion of the latitudinal correction to the model – observation difference, and (3) the use of a boundary condition correction. For each of these base inversions, we choose combinations of relative prior errors (50%, 75%, or 100%) and the regularization factor (between 0.001 and 1.0) so that the prior term of the cost function evaluated at the posterior solution averages to 1 across all grid cells where the averaging kernel sensitivities are greater than 0.05. We also require that at least 90% of these optimized grid cells are positive and that at least one grid cell is optimized for each of the 2386 model runs. This yields between one and three ensemble members for each of the base inversions, for a total of 15 members. We report the ensemble mean emissions, with uncertainty ranges given by the ensemble range.

**2.8 Source attribution**

The high resolution of the inversion facilitates the attribution of the posterior emissions estimates to different source categories, including sectors, states, and urban areas. We aggregate the native resolution emissions estimate and associated errors to find the corresponding quantities for each source category using a summation matrix , where is the number of source categories. The rows of are given by the relative contribution of each grid cell to a given source category. For sectoral attribution, the rows are given by the relative contribution of each sector to a grid cell in the prior emissions estimate. For state and urban area attribution, the rows are given by the fraction of each grid cell within the state or urban area, respectively. Urban areas are defined using the 380 U.S. Census 2019 Metropolitan Statistical Areas, which consist of the county or counties with at least one urban core with a population greater than 5104 and any surrounding counties with a high degree of commuting into the core. The reduced-dimension posterior estimate and associated posterior covariance and averaging kernel matrices are then given by

where is the Moore-Penrose pseudo inverse (Caliesi et al., 2005).

This approach to source attribution assumes that the distribution of emissions in the prior is correct and that emission sources are evenly distributed in grid cells that cross state or urban lines. Alternative approaches use the posterior error standard deviations to attribute the posterior emissions to sources, thereby including information from the observing system (Shen et al., xx; Cusworth et al, xx.). We use the simpler approach because the high resolution of the inversion decreases the chance of source co-location or significant distributional errors across boundaries.

**3 Results and discussion**

Figure 3 shows the ensemble mean posterior scale factors relative to the prior emissions estimate (left) and ensemble mean averaging kernel sensitivities (right). Boundary condition optimization results are shown in table 2. We find 784 (338 - 1278) DOFS across the domain, where the values in parentheses are the ensemble minimum and maximum, respectively. Of these DOFS, 38 (11 - 69) are found in Canada, 648 (281 – 1058) in the contiguous United States (CONUS), and 89 (45 - 134) in Mexico. The spatial variation in information content results from variations in prior emissions (e.g., small total emissions in Northern Mexico as shown in Figure 1) and in the observation density (e.g., low TROPOMI coverage north of 50 degrees latitude as shown in Figure 2). Nonetheless, we find a large increase in information content relative to past inversions over the same domain: Lu et al. (2022) found 114 DOFS in a joint inversion of data from GOSAT and NOAA’s ObsPack, while Shen et al. (2022) found 201 DOFS in an inversion of TROPOMI observations over 14 oil and natural gas basins. This increase reflects both the improved constraint provided by TROPOMI and the benefit of achieving high resolution on continental scales.

We use the averaging kernel sensitivities greater than 0.05 to define the grid cells that are optimized by the reduced-rank inversion. Optimized grid cells explain 21% (11% - 32%) of prior emissions in Canada, 74% (60% - 83%) in CONUS, and 66% (53% - 71%) in Mexico, reflecting the spatially variable information content. Across the optimized grid cells, we find a mean of 258 (40 - 572) negative values out of the 23691 grid cells optimized by the inversion across the ensemble. These values are mostly small and of the same order of magnitude as the soil sink.

We conduct an independent evaluation of the reduced-rank inversion by comparing our mean posterior emissions to a full-rank regional inversion of TROPOMI observations to quantify emissions from major oil and natural gas producing regions (Shen et al., 2022). [Insert]

The mean posterior emissions when used in GEOS-Chem decreases the root mean squared error of the simulated observations with the TROPOMI data by xx% relative to the prior GEOS-Chem simulation. [Insert posterior evaluation.] We also compare the prior and mean posterior simulations to in situ surface and aircraft observations from the GLOBALVIEWplus CH4 ObsPack v(xx) database maintained by the National Oceanic and Atmospheric Administration (NOAA) Global Monitoring Laboratory (cite). [Insert independent posterior evaluation.]

**3.1 National sectoral emissions**

Across the inversion ensemble, we find that relative to the prior, mean posterior anthropogenic and natural methane emissions decrease -0.8 (-2.2 - 0.1) Tg a-1 in Canada, increase 3.1 (1.5 - 4.7) Tg a-1 in CONUS, and decrease -0.7 (-1.3 - 0.1) Tg a-1 in Mexico. The indeterminate sign of the emissions change in Canada and Mexico results from the low fraction of prior emissions optimized by the reduced-rank inversion, which reflects the low information content available in these regions. As a result, we focus our discussion on CONUS.

We find posterior anthropogenic emissions of 31.4 (30.0 - 33.0) Tg a-1 in CONUS. This represents a 9% (5% - 15%) increase from our prior estimate of 28.7 Tg a-1 and a 17% (12% - 23%) increase from the most recent EPA Greenhouse Gas Inventory (GHGI) estimate of 26.8 Tg a-1 for 2019. Lu et al. (2022) found lower anthropogenic methane emissions of 28.9 (28.4 - 29.4) Tg a-1 over the same domain for 2017 by optimizing emissions and trends in a joint inversion of GOSAT and in situ observations for 2010 - 2017. Deng et al. (2022) conducted a survey of GOSAT inversions and found similar median posterior anthropogenic methane emissions for the United States of 26.5 (20.8 – 38.7) Tg a-1 for 2019. The lower estimate in these inversions could reflect changes between 2017 and 2019, different prior emissions, or differences between GOSAT and TROPOMI. Notably, GOSAT has similar accuracy and precision to TROPOMI, but produces two orders of magnitude fewer measurements. Deng et al. (2022) also surveyed inversions of in situ data, which used high accuracy and high precision observations from surface sites across the United States. In this case, they found median posterior anthropogenic methane emissions of 31.9 (23.9 - 43.1) Tg a-1, which closely matches our result and suggests that the lower estimate results at least partly from TROPOMI’s improved observational density. This explanation is further supported by the increase in DOFS we find compared to Lu et al. (2022) and by the narrower posterior emissions ranges we find compared to both Lu et al. (2022) and both inversion ensembles in Deng et al. (2022).

We allocate our national total to the emission sectors described in section 2.2 using the attribution method described in section 2.8. We find very low posterior error correlation between the sectors (mean error correlation coefficients less than 0.2 in all cases), indicating that we can successfully separate sectoral emissions. Figure 4 shows the resulting prior and mean posterior emissions for each sector in Canada, CONUS, and Mexico and table 1 shows the same data for CONUS. Almost all significant changes in sectoral emissions are observed in CONUS, with the largest changes occurring for the biggest anthropogenic methane emission sources, including landfills, livestock, oil and natural gas, and coal. We find little adjustment to wastewater, wetlands, and other sources in part due to the observing system’s inability to correct for regions containing these sources. For the sectors with significant adjustments, we find sectoral averaging kernel sensitivities between 0.49 and 0.92, with an average increase of 64% relative to the equivalent values found by Lu et al. (2022), indicating that TROPOMI provides an improved constraint on sectoral emissions. With the exception of landfill emissions, we also find a narrower ensemble spread than Lu et al. (2022). Each sector is also well constrained by the reduced-rank inversion: on average, between 65% and 81% of the prior sectoral emissions are optimized.

Landfill emissions show the largest relative increase from the prior emissions estimate. We optimize 81% (62% - 90%) of prior landfills emissions and find a sectoral averaging kernel sensitivity of 0.49 (0.31 – 0.66), indicating that on average the observing system informs 49% of the posterior estimate with the remainder provided by the prior. We find posterior emissions of 7.2 (6.4 - 8.2) Tg a-1, an increase of 27% (13% - 45%) relative to the prior and 58% (41% - 81%) relative to the most recent EPA GHGI estimate for 2019. Lu et al. (2022) found similar mean posterior landfill emissions of 7.2 (6.0 - 7.6) Tg a-1 for 2010 – 2017 with a sectoral averaging kernel sensitivity of 0.34. [Apply trends analysis] We attribute the increase to underestimated urban emissions, including both direct landfill emissions and co-located post-meter natural gas emissions, which we discuss in section 3.3.

Livestock emissions show the largest absolute increase from the prior emissions estimate. We optimize 65% (45% - 76%) of prior emissions and find a sectoral averaging kernel sensitivity of 0.67 (0.50 - 0.77). Posterior emissions of 10.5 (9.9 - 10.9) Tg a-1 represent a increase of 14% (8% - 19%) from the prior and 11% (5% - 16%) from the most recent EPA GHGI estimate for 2019. Lu et al. (2022) found similar mean posterior livestock emissions of 10.6 (9.2 - 11.8) Tg a-1 over CONUS for 2010 – 2017 with a sectoral averaging kernel sensitivity of 0.43. [Apply trends analysis] Yu et al. (2021) conducted a seasonal inversion of aircraft observations over the north central United States and south central Canada to find mean posterior livestock emissions of 5.5 (5.1 - 6.2) Tg a-1, which agrees well with our posterior livestock estimate of 5.4 (5.1 - 5.6) Tg a-1 over the same region. We attribute the low EPA GHGI estimate to underestimated manure management emissions, which we discuss in section 3.4.

Our posterior emissions for fossil fuel emissions show a significant increase in information content produced by the improved observational density offered by TROPOMI. We optimize 88% (80% - 92%) of prior oil and natural gas emissions and almost all prior coal mining emissions, and our sectoral averaging kernel sensitivities are twice as large as those found in Lu et al. (2022). Posterior oil and natural gas emissions are 10.5 (10.2 - 10.9) Tg a-1, an increase of 11% (8% - 15%) from the prior emissions estimate and 15% (12% - 20%) from the most recent EPA GHGI estimate for 2019. The larger discrepancy from the updated EPA GHGI estimate reflects the inventory’s decreased production emissions despite increases in processing, transmission, and storage emissions and the addition of post-meter emissions. Coal mining emissions exhibit the largest decline in sectoral emissions, decreasing 49% (34% - 61%) relative to the prior and 30% (10% - 46%) relative to the most recent EPA GHGI estimate for 2019. The decrease in emissions is consistent the 30% decrease in CONUS coal production since 2012 (USGS).

We also find broad consistency with past studies of fossil fuel methane emissions. Lu et al. (2022) found much larger fossil fuel posterior emissions of 4.6 (3.0 - 4.7) Tg a-1 for oil, 9.9 (8.1 - 10.5) Tg a-1 for natural gas, and 2.8 (2.4 - 3.5) Tg a-1 for coal for 2010 - 2017. However, they found decreasing natural gas emissions from 2014 to 2017 that could explain part of the discrepancy if they continued until 2019. Shen et al. (2022) found slightly lower oil and natural gas emissions of 12.6 ± 2.1 Tg a-1 from an inversion of TROPOMI data over oil and natural gas basins extrapolated to the national scale for May 2018 to 2020, which is consistent with continued emissions decreases. Deng et al. (2022) found much smaller cumulative fossil fuel emissions of 9.8 (8.1 - 13.7) Tg a-1 for GOSAT inversions and 12.6 (8.0 - 16.7) Tg a-1 for in situ inversions in 2019, both of which are consistent with our result.

**3.2 State sectoral emissions**

The high resolution achieved by our inversion allows for finer-scale source attribution. Within CONUS, we partition emissions, excluding offshore emissions, to each of the 48 states as described in section 2.8. Figure 5 shows state prior and posterior emissions, table 4 shows the corresponding data for the top 10 methane producing states, and table S1 shows the full results. TROPOMI provides a strong constraint at this resolution, with mean state averaging kernel sensitivities larger than 0.5 in 29 of all 48 states and in 20 of the top 25 methane producing states. Our reduced-rank inversion also constrains emissions in most states: we optimize an average of 80% of prior emissions in each of the top 25 methane producing states.

Emissions are consolidated in the largest methane-producing states, with 8 states responsible for 50% of mean posterior emissions in CONUS (7 for prior emissions) and 7 states responsible for more than 54% of inferred emissions increases. Texas alone produces 21% of the mean posterior emissions (18% in the prior) and 25% of the inferred emissions increase. Oil and natural gas emissions account for 66% of the observed increase in Texas and are concentrated over the Permian basin, the largest oil and natural gas producing region in CONUS (cite). We also find significant increases over the Texan portions of the Haynesville shale and over the Houston metropolitan area. More generally, we find significant emission increases in 7 of the top 10 methane producing states, with a significant decrease in New Mexico and no significant change in Pennsylvania and Kansas. Oil and natural gas are responsible for 47% of mean posterior emissions and 44% of the observed increase in these 10 states, compared to the national average of 33% and 36%, respectively.

Shifting trends in methane emission sources since 2012 result in a significant re-ordering in the largest methane-producing states from the prior to the posterior. The largest increases in ranking occur in Florida (21st to 8th), Indiana (26th to 15th), Georgia (29th to 19th), and Mississippi (33rd to 24th). Of these, Indiana, Georgia, and Mississippi are sensitive to the observing system, with mean state averaging kernel sensitivities between 0.5 and 0.6, while Florida has a mean sensitivity of 0.32. In each of these three states, the observed ranking increase is driven by increased landfill emissions. The largest decreases in emissions rank all occur in coal-producing states, including Wyoming (14th to 42nd), West Virginia (5th to 20th), Kentucky (17th to 27th), and Virginia (19th to 28th), all of which have state averaging kernel sensitivities greater than 0.5. This reordering reflects a systematic underestimate of urban emissions (section 3.2) and a shift in the dominance of methane emission sources in the last decade.

We compare our posterior emissions to the EPA state emissions inventories for 2019, scaled to match the total mean posterior emissions. On average, our mean posterior emissions are only 4% smaller than the scaled EPA estimates. However, the distribution of the posterior – inventory differences is highly skewed, with the largest relative discrepancies found in the largest methane producing states: in the top 10 methane producing states, we find that the state posteriors are on average 18% larger than the scaled EPA estimates. Consistent with our sectoral analysis, the largest absolute negative posterior – inventory differences are found in historically coal-producing states, including Wyoming, Pennsylvania, and West Virginia. The largest positive differences occur in states with large oil and natural gas emissions, including Texas, Louisiana, and New Mexico.

We also compare our posterior emissions to state greenhouse gas inventories for the year closest to 2019 where available and where the observing system provides a strong constraint (state averaging kernel sensitivity greater than 0.5). Of the inventories of California, Colorado, Iowa, Louisiana, and Pennsylvania, our posterior agrees only with Pennsylvania. While posterior emissions are not significantly different from the prior in Pennsylvania, we find a source shift from fossil fuels (from 73% in the prior and 76% in the inventory to 63% in the posterior) to landfills and livestock (from 25% in the prior to 34% in the posterior). The inventories of California, Iowa, and Louisiana are smaller than our mean posterior emissions, while Colorado finds larger emissions, with different driving sectors in each state. Our prior and mean posterior estimates are larger than the California inventory by 13% and 32%, respectively, with 78% of the inferred increase attributable to livestock and 14% to oil and natural gas. In Iowa, our prior estimate is only 4% smaller than the inventory value, but we find a mean 33% increase in emissions that is attributed to livestock (70%) and landfills (18%). The Louisiana inventory finds methane emissions half as small as our prior, and we find an additional mean 65% increase in emissions attributed to oil and natural gas (71%) and other sources, predominantly rice (16%). The Colorado inventory is larger by 44% than our prior estimate and 65% than our mean posterior emissions. The difference between the inventory and the inversion is due to oil and natural gas, where the inventory estimates emissions more than twice as large as our prior.

**3.3 Urban sectoral emissions**

Urban areas, as defined by the U.S. Census Bureau’s metropolitan statistical areas (MSAs), are disproportionately responsible for methane emissions in CONUS. We calculate the mean posterior emissions for the 351 metropolitan statistical areas (MSAs) defined by the U.S. Census Bureau (section 2.8) that are optimized by at least one of the 15 inversion ensemble members. These urban areas occupy 30% of CONUS surface area but are responsible for 41% of prior emissions. The observing system provides a weaker constraint on urban areas than on sectors or states due to the larger TROPOMI errors incurred by the urban grid and due to smoothing errors, with mean urban averaging kernel sensitivity values of 0.24 (0.12 - 0.35). We still find significant corrections: posterior emissions are on average 9% (20% - 33%) larger than the prior.

The observed increase in urban emissions suggests an underestimate of landfill emissions or natural gas distribution or post-meter emissions. The EPA added post-meter emissions of 456 Gg a-1 to their most recent GHGI for 2019, representing 5% of oil and natural gas emissions. We find a net adjustment of 2592 (1396 - 4038) Gg a-1 to urban areas, of which 18% (11% - 33%) is explained by the EPA post-meter emissions. In most urban areas, we find that we are unable to explain the remaining discrepancy because source co-location results in high sectoral covariance. We also find no correlations between the observed increase in either aggregated urban areas or individual urban grid cells and urban area population, population change from 2012 to 2019, population density, or surface area. The lack of correlation reflects the variability of methane emission sources and trends between urban areas.

We also

Top 15 cities

New York

LA

Chicago

Dallas

Houston

DC

Miami

Philadelphia

Atlanta

Phoenix

Boston

SF

Riverside

Detroit

Minneapolis

Compare to city inventories

Compare to other studies of urban areas

**3.4 Livestock emissions**

[Insert work on identifying the cause of the livestock increase.]

65% (45% - 76%) of prior livestock emissions

EPA GHGI 9.4 total

EPA GHGI enteric fermentation 7.0

EPA GHGI manure management 2.3

**4 Conclusions**

[Insert.]