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

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We use 2019 atmospheric methane columns measured by the Tropospheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5 Precursor satellite in an inversion that quantifies methane emissions at 0.25° ⨉ 0.3125° resolution over North America. We perform substantial cleaning of the TROPOMI data to avoid regional biases. We solve the inversion by analytical minimization of a Bayesian cost function, which provides closed-form expressions for the error and information content of the inversion and allows for the creation of an ensemble of inversions to test the sensitivity of different uncertainties and assumptions. We achieve high resolution results across North America with a reduced-rank eigenvector characterization of the observing system that maximizes information content while managing computational cost. Prior methane emission estimates for the inversion are gridded versions of the national inventories reported by individual countries under the Paris Agreement. We use inversion results to correct the methane emissions in these national inventories for different sectors and regions, providing a top-down framework for using satellite observations to improve national methane emission reporting.

**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). In the case of a linear relationship between emissions and concentrations, the optimal (posterior) solution is typically found by numerically or analytically minimizing a Bayesian cost function regularized by a prior emissions estimate. The numerical (variational) approach uses the adjoint of the CTM together with gradient descent algorithms. 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 2.4 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.

**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).

The standard analytical solution is numerically unstable for large since it requires inverting a non-sparse matrix. We instead use reduced-rank approximations that solve the inversion on 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 the matrix of the first eigenvalues. We choose = 1952 to match the rank of the Jacobian matrix.

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

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. The reduced-rank Jacobian matrix (section 2.6) optimizes emissions only in those grid cells that are informed by the observing system and defaults to the prior elsewhere. We also optimize 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). 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. We use the high-performance WetCHARTs ensemble version 1.3.1, which selects from an ensemble of process-based models the 10 that best agree with Greenhouse gases Observing SATellite (GOSAT, described section 2.4) observations (Ma et al. 2021). We decrease 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 (cite). We also conduct a sensitivity test where we use the unscaled WetCHARTs emissions but eliminate two ensemble members that produce anomalously large emissions in the high northern and southern latitudes in summer and fall (Lu et al. 202?). Other natural methane emission sources include open fires, termites, and geological seeps. Open fire emissions are from the Global Fire Emissions Database version 4 (GFED4, van der Werf et al., 2017), termite emissions from Fung et al. (1991), and geological seepage from Etiope et al. (2019) scaled to the 2 Tg a-1 global emission magnitude given by Hmiel et al. (2020).

We assume uniform relative prior errors of xx% following previous inversions that optimized methane emissions over North America (Maasakkers et al., 2021; Lu et al., 2022). In the absence of better information, we assume there is no error covariance. We conduct sensitivity tests with variable uniform relative errors and using sector-specific errors from the EPA GHGI.

**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 observations used provide significant coverage only between 30°N and 50°N. We expect no systematic bias with respect to albedo because GEOS-Chem does not simulate radiative transfer.

**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 (section 2.2).

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.

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 the first-order polynomial

where is the degrees latitude. We conduct a sensitivity test where we do not apply the latitudinal correction.

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 = xx). 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). While filtering improves the performance of the TROPOMI data relative to GOSAT and to the prior GEOS-Chem simulation, 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 (section 2.5).

**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 account for 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 820863 (28%) observations. We find a mean observational error standard deviation of 11.64 ppb, with larger errors in winter and at high latitudes. We take the corresponding variances as 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 = xx (section 2.1) to account for this lack of covariance (Chevallier et al., 2007). We determine by

**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 (Nesser et al., 2021).

**3 Results and discussion**

Figure 3 (right) shows the averaging kernel sensitivities greater than or equal to 0.05 for the base inversion. These averaging kernel sensitivities show where the reduced-rank observing system constrains the emissions estimate (Nesser et al., 2021). We find a total of 807 DOFS, 710 of which occur in the 4578 grid cells with averaging kernel sensitivities greater than or equal to 0.05. These grid cells explain 73% of prior emissions in CONUS (558 DOFS), 68% in Mexico (88 DOFS), and 30% in Canada (56 DOFS). The discrepancies in coverage result 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., little TROPOMI coverage north of 50 degrees latitude as shown in Figure 2). Due to the lack of observing system constraint in Canada, we focus our discussion on CONUS and Mexico. Over these domains, we find a large increase in the observing system constraint 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 across the domain.

Figure 3 (left) shows the posterior scaling factors relative to the prior emissions and Table 1 summarizes the sectoral results from CONUS and Mexico.

Figure 4 shows

* Summary paragraph for total U.S., Canada, and Mexico emissions results
* Paragraph on posterior evaluation (posterior concentrations vs. TROPOMI, posterior concentrations vs. surface/aircraft)
* Go through sectoral results:
  + Top-line summary paragraph (describing the prior/posterior bar graph figure, including error bars, and describing sectoral averaging kernel values).
  + Oil and natural gas, including a basin-by-basin analysis using Lu's basins (and expanding to other regions included in our state vector that were excluded by Lu, e.g. the Saskatchewan oil sands). We'll also need to explain the different results in Mexico and in the Marcellus.
  + Coal, since we find such a large decrease that is consistent with EPA trends (and it may explain the Marcellus result)
  + Livestock, including (if interesting) a breakdown by manure management vs. enteric fermentation, since I think a lot of our corrections may correspond with manure management
  + Wetlands
* Other interesting results:
  + States
  + Cities

**4 Conclusions**