**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 modelled pathways that limit global warming to 1.5°C require deep reductions in methane emissions (IPCC). Yet, global methane concentrations have tripled from pre-industrial concentrations and are increasing at an increasing rate (Dlugokencky). Methane is emitted by a range of sectors, including the fossil fuel industry, livestock, waste management, and wetlands, the main biogenic source. Most countries, including Canada, the United States, and Mexico, report their total estimated anthropogenic methane emissions to the United Nations Framework Convention on Climate Change (UNFCCC) using methods defined by the International Panel on Climate Change (IPCC). These “bottom-up” methods combine activity data (e.g., number of natural gas pipelines) with emission factors (e.g., the leakage rate per pipeline) to estimate total emissions. However, significant uncertainties exist in both the spatial and temporal variability of emission factors, leading to large uncertainties in total and sectoral methane emission estimates. Satellite observations of atmospheric methane concentrations can improve bottom-up estimates with increasing sectoral accuracy through high-resolution inverse analyses (Streets et al. 2013, Jacob et al. 2016). We evaluate gridded versions of the national inventories of Canada, the United States, and Mexico for 2019 using observations of column methane concentrations from the TROPOspheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5 Precursor. We estimate methane emissions at 0.25° ⨉ 0.3125° resolution, allowing for improved hotspot identification and source attribution.

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 x 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° x 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 the TROPOMI observations and the observing system errors; and section 2.4 describes GEOS-Chem and the novel, reduced-rank method used to calculate the Jacobian matrix. We also conduct a suite of sensitivity tests to provide additional constraints on the error of the optimized emissions, which are summarized in section 2.5.

**2.1 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, respectively (section 2.4); and are the observational vector and error covariance, 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). The nested methane CTM is linear so that where is the Jacobian matrix and is constant, allowing analytical minimization of the cost function yielding the optimal (posterior) state vector estimate and associated error covariance matrix :

The relative reduction in the error covariance defines the information content of the observing system, quantified by the averaging kernel matrix

The averaging kernel matrix describes the sensitivity of the posterior estimate to the true state vector, . The diagonal elements are therefore 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).

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

We optimize emissions in 23,691 grid cells at 0.25° x 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. Methane chemical and soil sinks are not optimized because these loss processes are slow compared to the ventilation timescale.

Table 1 summarizes the prior emissions estimates and Figure xx shows the spatial distribution of major source sectors. 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. 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 find that two of the ensemble members produce large methane emissions inconsistent with previous inversions of GOSAT data in the high northern and southern latitudes in summer and fall (Lu et al. 202?). We conduct a sensitivity test that eliminates those ensemble members. We also test the effect of decreasing total wetland emissions by a factor of 4.04, which is derived from a comparison of the ensemble to FLUXNET CH4, a network of eddy covariance tower data (cite).

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 errors of 100%. Previous inversions that optimized methane emissions over North America used 50% relative errors (Maasakkers et al., 2021; Lu et al., 2022, p.2). However, these inversions optimized emissions using ~600 Gaussian basis functions of varying resolution. Because we optimize emissions at high resolution everywhere where the observing system has information, we inflate the errors. We conduct a sensitivity

We conduct a sensitivity test using sectorally variable errors from the EPA GHGI. We determine the …

In the absence of better information, we assume there is no error covariance. However, this assumption is worse than

**2.3 Forward model**

We use the nested version of the GEOS-Chem chemical transport model (CTM) v12.7 at 0.25° x 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 (qa = 1).

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° x 2° grid following Lorente et al. (2021). We find large regional biases, defined as the standard deviation of the 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 disproportionately account for the remaining unphysical TROPOMI observations (XCH4 < 1700 ppb) and exhibit large prior GEOS-Chem – TROPOMI biases. Finally, we find anomalous prior GEOS-Chem – TROPOMI differences north of 50°N in winter. We remove these scenes since they are likely to correspond with snow- and ice-cover. The resulting 2919358 observations preserve 69% of the original high-quality data and decreases seasonal regional biases by between 7% and 21%. In all seasons, the regional biases are 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).

Figure xx (solid lines) shows the prior GEOS-Chem – TROPOMI difference with respect to albedo, season, and latitude for the filtered data. We find no bias with respect to albedo or season and an aseasonal latitudinal bias. 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 find good agreement between the resulting prior GEOS-Chem output and the observations (R = 0.77).

Figure xx (top row) shows the final observations, regridded onto the GEOS-Chem grid and averaged seasonally, that constitute our observation vector **y**. The observational density (middle row) illustrates the heterogenous coverage of the data, which is the basis for the reduced-rank approach we take to construct the Jacobian matrix for the inversion (Section 2.6). While filtering improves the performance of the TROPOMI data relative to GOSAT and to the prior GEOS-Chem simulation, we still find large seasonal gradients in the prior GEOS-Chem – TROPOMI difference (e.g., in spring over Northern Wisconsin), as shown by the bottom row of figure xx. These differences may be due to errors in the prior, but the steepness of the gradient suggests the possibility of residual systematic biases in the observations. We account for these biases in our error specification (section 2.5).

**2.5 Observing system errors**

The bottom row of figure xx shows the error standard deviations of the observing system, including contributions from the forward model, the instrument, and representation error, regridded onto the GEOS-Chem grid and averaged seasonally. 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 and month is caused by errors in emissions that will be corrected by the inversion. The residual error after subtracting the mean seasonal, gridded error gives an estimate of the observational error. In the 1% of 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 We find a mean observational error standard deviation of 9.5 ppb that does not vary significantly with albedo, season, or latitude. 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.

*[I realize now that given that we are optimizing emissions annually, perhaps the mean errors should be calculated for each grid box, but not for each month.]*

We introduce a regularization factor (section 2.1) to account for this lack of covariance. Chevallier et al. (2007) showed that error inflation could account for unquantified covariance in the observing system error covariance matrix. We draw on the parallel use of the prior and observing system error covariance matrices *[Isn’t there a source that says that scaling up the diagonal produces equivalent results to including off-diagonal elements?]* ….

We introduce a regularization factor (section 2.1) to account for the lack of covariance structure in . *[Isn’t there a source that says that scaling up the diagonal produces equivalent results to including off-diagonal elements?]* ….

Houweling et al. (2017):

“Errors that behave quasi-random and affect neighboring retrievals in a coherent way can in theory be accounted for by specifying the off-diagonal terms in the data error covariance matrix. In practice, there are many ways to do this, but quantitative information to justify a specific choice is lacking. In general, correlated uncertainty reduces the number of independent measurements, which justifies averaging retrievals within a certain distance of each other. Usually the uncertainty of the mean is calculated using a lower bound representing the contribution of purely systematic error. An alternative approach, referred to as ”error inflation”, is to increase the error of individually assimilated retrievals such that the uncertainty of a mean of surrounding retrievals does not drop below this minimum level (Chevallier, 2007). The advantage of this approach is that it avoids subjective decisions about which samples to combine into an average. Error inflation, or similar methods that compensate the neglect of off diagonals in the data error covariance matrix by increasing the (diagonal) uncertainty, lead to a χ 2 below 1. Although this may seem suboptimal from a statistical point of view, Chevallier (2007) demonstrated that this de-weighing of data nevertheless leads to uncertainty reductions that are closer to those obtained when off diagonals in R had been accounted for. Therefore, this approach avoids over constraining the problem by neglecting the contribution of data error covariance.”

“Observational error covariances are prescribed as the relative residual standard deviation of the column mismatch between the true-state synthetic observations and the prior simulations over a 2◦ × 2 ◦ moving window (Heald et al., 2004). We impose on the derived values a lower limit of 60 ppb2 , corresponding to the 0.25 quantile of the overall error distribution. The resulting observing system errors average 9 ppb (range: 8–29 ppb) and mainly reflect instrument noise. The 9 ppb estimate is in line with and slightly smaller than observational error estimates for previous methane inversions using data from TROPOMI (e.g., 11 ppb; Zhang et al., 2020) and GOSAT (e.g., 13 ppb; Maasakkers et al., 2019); it is therefore an appropriate representation for our OSSE analyses. Note that any systematic measurement errors (Lorente et al., 2021) are inherently not accounted for in our framework and would need separate correction.” (Yu et al. 2021)

**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 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 (equation 4) 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 patterns of information content are then given by the eigenvectors of the averaging kernel matrix described by the columns of

where is the matrix of the first *k* eigenvectors of .

We then construct the Jacobian matrix on the basis of these eigenvectors. We select the eigenvectors that have at least twice as much signal as noise and perturb those patterns in the forward model. We multiply the resulting Jacobian matrix by the matrix

to restore the original state dimension, yielding an updated Jacobian matrix estimate . We then recompute the eigenvectors using perturb the eigenvectors that explain xx% of the information content, and construct the updated Jacobian matrix . This update scheme follows Nesser et al. (2021), which found that the information content of the inversion was more sensitive to perturbation simulations in the second update. The information content associated with both and contains common contributions from the prior and observational error covariance matrices and from the forward model. As a result, additional iterations would not add new information content and we take as the Jacobian matrix for our inversion.

[Paragraph analyzing the eigenvectors of the resulting Jacobian matrix.]

**3 Results and discussion**

“This reflects a tendency for SF inversions to overcorrect large sources while undercorrecting small sources (along with the fact that the satellite data themselves are less sensitive to small sources).” (Yu et al. 2021)

🡪 ask Daniel about this

“The U-SF inversion has DOFS = 382, with derived posterior error reductions that reflect the TROPOMI spatial sampling density for this month (Fig. 7). However, this computed error reduction ρest (derived via gradient-based randomization) has no meaningful spatial correlation with the actual emission improvement ρtrue (R = 0.07). This reflects the fact that the posterior error reductions and DOFS contain no information on where the prior emissions are actually in error and can therefore be improved. For a scenario where the prior emissions had random and normally distributed disparities relative to the truth, areas with the largest computed posterior error reduction would also tend to have the greatest emission improvement – since those locations would have the strongest observational constraints. DOFS and error reduction analyses are thus useful for general observing system characterization but do not describe the spatial accuracy of posterior emissions or the actual emission improvements for realistic scenarios where the real prior errors are nonrandom.”

🡪 ask Daniel about this

Often see better performance for small sources than large sources, since prior errors are probably more appropriate for small sources than for large sources (can cite Yu et al. to explain why Permian estimates may be too low)

**4 Conclusions**