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

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We quantify 2019 methane emissions in the contiguous United States (CONUS) at 0.25° × 0.3125° resolution by inverse analysis of atmospheric methane columns measured by the Tropospheric Monitoring Instrument (TROPOMI). A gridded version of the U.S. Environmental Protection Agency (EPA) methane inventory reported under the Paris Agreement serves as the prior estimates for the inversion. We optimize emissions and quantify observing system information content through analytical minimization of a Bayesian cost function. We achieve high resolution with a reduced-rank characterization of the observing system that preserves information content by optimizing emissions only where observing system errors are low or prior emissions are large. The resulting inversion decreases computational cost by an order of magnitude while optimizing 80% of anthropogenic emissions in CONUS. We find mean posterior anthropogenic emissions of 30.9 (30.0 - 31.8) Tg a-1, where the parenthetical values give the spread of an 8 member inversion ensemble, a 15% increase from the most recent EPA Greenhouse Gas Inventory (GHGI) estimate for 2019 of 26.8 Tg a-1. Mean livestock emissions are 10.4 Tg a-1, oil and natural gas 10.4 Tg a-1, coal 1.5 Tg a-1, and landfills 6.9 Tg a-1. We find a 52% increase and 30% decrease from the GHGI for landfills and coal, which we attribute to underestimated urban emissions and decreasing coal production, respectively. The GHGI livestock emissions increase of 11% correlates with hog and dairy cattle operations, suggesting underestimated manure management emissions. We partition our emissions to the 48 states and compare to EPA’s new state inventories. Posterior emissions are on average 38% larger than these inventories in the largest 10 methane-producing states. We find the largest upward adjustments in states with large oil and natural gas emissions and the largest downward changes in states with large contributions from coal emissions. We also calculate emissions for 219 geographically diverse urban areas. Although these urban areas occupy only 2.2% of CONUS surface area, posterior emissions explain 8% of the national total, equivalent to almost quarter of oil and natural gas emissions. Relative to the gridded EPA emissions, we find a 29% upward adjustment with a 69% increase in the largest 10 methane-producing cities. The addition of post-meter natural gas emissions to the GHGI could explain 82% of the adjustment, but variability across urban areas hinders the attribution of the remainder of the discrepancy. Our ability to quantify point and area sources demonstrates the value of achieving high resolution at continental scales and lays the groundwork to improve the sectoral, national, and regional attribution of greenhouse gas emissions inferred from satellites.

**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 emission inventories use information on sectoral activity levels and emission factors, but considerable uncertainty can exist in these values. Top-down evaluations of bottom-up inventories use an atmospheric transport model to relate observations of atmospheric methane to emissions. Accurate evaluation requires optimizing emissions at high spatial resolution and characterizing the error and information content of the observation-model 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° (≈25 × 30 km2) resolution over North America for 2019, allowing detailed analysis of sectoral, state, and urban emissions.

Satellite observations of atmospheric methane column concentrations inferred from measurement of backscattered sunlight in the shortwave infrared have been extensively used in inverse analyses of methane emissions (Streets et al., 2013; Jacob et al., 2022). Past satellite instruments were limited by low information content resulting from large pixel size (SCIAMCHY, 2003 – 2012) or sparse observations (GOSAT, 2009 – present). The Tropospheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5 Precursor satellite now provides daily, global observations of atmospheric methane columns at 5.5 km × 7 km nadir pixel resolution over land (Hu et al., 2018; Lorente et al., 2021) with a 3% success rate limited by cloud cover, optically dark surfaces, and heterogeneous terrain (Hasekamp et al., 2019). Inversions of TROPOMI data allow for high-resolution emission quantification but require understanding the actual information content of the observations.

Inverse analyses optimize methane emissions (the state vector) by fitting observations to simulated concentrations from a chemical transport model (CTM). This is typically done by minimizing a Bayesian cost function regularized by a prior emissions estimate given by a bottom-up inventory. When a linear relationship exists between emissions and concentrations, the optimal (posterior) solution and the associated errors and information content can be found analytically. This method also supports the generation of inversion ensembles. However, it requires explicit computation of the Jacobian matrix that represents the relationship between emissions and concentrations in the CTM, which is typically constructed through finite difference by conducting a perturbation simulation for each emission element. This limits the resolution of the optimized emissions. Past analytical inversions achieved high resolution by regridding coarse-resolution results (e.g., Cusworth et al., 2021), by aggregating grid cells (e.g., Turner et al., 2015; Maasakkers et al., 2021), or by limiting the optimized domain (e.g., Yu et al., 2021; Shen et al., 2022; Plant et al., 2022). These approaches relied on prior knowledge. Nesser et al. (2021) integrated information from the observing system by constructing the Jacobian matrix on the basis of the emissions patterns best informed by both the bottom-up inventory and the observations. This approach optimally preserves the information content of the observing system by optimizing emissions at high resolution only where the observing system provides a constraint and defaulting to the prior elsewhere.

Several inverse studies have found large national discrepancies between bottom-up emissions estimates from the U.S. Environmental Protection Agency’s Greenhouse Gas Inventory (GHGI) and inferred emissions from satellite, aircraft, and in situ observations. Lu et al. (2022) found mean 2010 – 2017 anthropogenic emissions of 36.0 (32.5 - 37.8) Tg a-1 compared to the 2021 EPA GHGI estimate of 26.0 Tg a-1 for the same period from an inversion of data from the Greenhouse Gases Observing Satellite (GOSAT), which they attributed largely to oil emissions over the south-central US. They also found increases in livestock (15%) and landfill (24%) emissions relative to the GHGI but were unable to explain the source of the increase, due in part to the coarse resolution of the inversion over those source regions. Deng et al. (2022) determined national methane emissions from the Global Carbon Project inversion ensemble (Saunois et al., 2020), and found median 2017 emissions of 26.5 (20.8 - 38.7) Tg a-1 for GOSAT inversions and 31.9 (23.9 - 43.1) Tg a-1 for inversions of in situ data. However, the large range prohibits accurate source attribution.

High resolution regional studies have worked to explain the origins of the observed national discrepancies. Alvarez et al. (2018) used aircraft and in situ observations from Shen et al. (2022) conducted an inversion of TROPOMI data over North American oil and natural gas basins from May 2018 to February 2020 and found emissions 80% larger than the EPA GHGI for 2021. Chen et al. (2018) and Yu et al. (2021) inverted in situ and aircraft observations over the upper Midwest and found an 80% and 24% increase, respectively, from the Gridded EPA (GEPA) inventory based on the 2016 EPA GHGI for 2012. Sheng et al. (2018) found good agreement with GEPA in an inversion of SEAC4RS aircraft observations over the Southeast US, but they found large regional variability. Plant et al. (2019) used aircraft observations of methane, CO2, and CO together with CO2 and CO emission inventories to infer methane emissions from six East Coast urban areas more than two times larger than GEPA. While these regional studies provide insight into some of the sources of the observed discrepancy with EPA estimates, they are hindered by their limited domain.

High-resolution inversions of TROPOMI satellite observations offer the opportunity to optimize national, regional, and sectoral methane emissions at high resolution. We used TROPOMI observations over North America for 2019 to optimize emissions and information content at 0.25° × 0.3125° resolution, sufficient to constrain sectoral, state, and urban emissions. Our prior emission estimate is provided by the GEPA inventory so that our corrections are policy relevant. We used cleaned TROPOMI data that avoids the largest biases with respect to albedo and snow- and ice-cover. We apply Nesser et al. (2021) to construct a reduced-rank Jacobian matrix that optimizes emissions only where the observing system provides a constraint and generate an 8-member, quality-controlled inversion ensemble that improves our estimation of errors. We partition our results by sector, state, and urban area, bridging our national total with local emissions.

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

We conduct an inversion of 2019 TROPOMI methane observations over North America (latitudes between 9.75° to 60° and longitudes between -130° and -60°, see domain shown in figure 1). The TROPOMI observations are fit to simulated concentrations from the GEOS-Chem 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 a reduced-rank approximation of the analytical minimum of a Bayesian cost function regularized by a prior estimate given by the U.S. EPA emissions reported under the Paris Agreement.

**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; and are the vector of observations and the associated error covariance matrix, respectively; and is the reduced-rank Jacobian matrix that represents the sensitivity of observations to emissions in the CTM as described in section 2.6 (Rodgers, 2000). The error covariance matrices are often assumed diagonal in the absence of better information, and is a regularization factor that accounts for the absence of covariance in (Chevallier et al., 2014). We generate an inversion ensemble using a range of prior errors, observations, and values (section 2.7).

Analytical minimization of the cost function 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 of are commonly referred to as averaging kernel sensitivities and their sum (trace of ) gives the degrees of freedom for signal (DOFS), the number of pieces of information independently constrained by the observing system (Rodgers, 2000). However, the reduced-rank Jacobian matrix prevents the computation of the standard posterior solution due to numerical instabilities in the inversion of a low-rank matrix required to calculate (Rodgers, 2000). We use instead 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 of the forward model

where the columns of are the eigenvectors and is a diagonal matrix with entries equal to the eigenvalues. The calculation of 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. The reduced-rank approximation is then generated using the largest eigenvalues and the associated eigenvectors :

Here, approximates the full-rank (FR) posterior by minimizing the difference between the two, and and are the optimal posterior error covariance and averaging kernel matrices for an inversion solved with a reduced-rank observing system. We choose to match the rank of the reduced-rank Jacobian matrix, which is chosen to maximize the DOFS within the available computational resources (section 2.8). To reduce noise generated by the reduced-rank inversion and reduced-rank Jacobian matrix, we filter out grid cells with averaging kernel sensitivities less than 0.05, leaving only “optimized grid cells.”

**2.2 Prior estimate and errors**

Figure 1 shows the sectoral spatial distribution of the prior emissions estimate. We evaluate the national inventories prepared under the Paris Agreement by using as anthropogenic emissions for the United States, Mexico, and Canada the spatially disaggregated (gridded) versions of the EPA GHGI for 2012 (GEPA; 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 (Shen et al., 2022). We also use the Environmental Defense Fund’s high-resolution inventory over the Permian basin, one of the largest oil and natural gas producing regions in North America, for 2019 (Zhang et al., 2020). We treat oil and natural gas as a single sector in our analysis due to significant source co-location and uncertainty in the partitioning of oil and gas wells. Anthropogenic emissions for Central America and the Caribbean islands are provided by the EDGAR v4.3.2 global emission inventory for 2012, though these represent only a small fraction of total emissions (?). 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 (Bloom et al., xxxx). We use the high-performance WetCHARTs ensemble version 1.3.1, which is a subset of the full WetCHARTs ensemble that performed best in comparisons to GOSAT observations (Ma et al. 2021). Lu et al. (2022) found a large overestimate of wetland methane emissions in the high-performance ensemble, particularly in the boreal wetlands. We remove from the high-performance ensemble two members that produce anomalously large emissions in the high northern latitudes in summer and fall. Other natural methane emission sources include open fires, termites, and geological seeps, for which we follow the emissions described in Lu et al. (2022).

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.

Across our inversion ensemble, we assume uniform relative prior error standard deviations for emissions of between 50% and 100%. 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 an error standard deviation of between 5 ppb and 10 ppb. We choose the prior error for each ensemble member as described in section 2.7. 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 CTM 12.7 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 1, 2019 and 3-hourly boundary conditions for the year are specified by methane concentration fields from a global GEOS-Chem simulation at 2° × 2.5° resolution using optimized emissions from a global inversion of TROPOMI observations (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. Initial conditions and boundary conditions are provided by the same simulation from Qu et al. (2021). 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**

TROPOMI 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). Methane concentrations are inferred from a full-physics retrieval that can fail due to cloud cover, variable topography, low or heterogeneous albedo, and high aerosol loading (citation). As a result, TROPOMI has a xx% retrieval success 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). GOSAT measures backscattered solar radiation at 1.6 μm with high-precision in 10 km diameter nadir pixels separated by ~250 km along- and cross-track. It has 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 their quality assessment. We compare average seasonal TROPOMI and GOSAT 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 20 ppb in winter, which is likely due to snow- and ice-cover. We remove these scenes using blended albedo > 0.75 in fall, winter, and spring (Wunch et al., 2011; Lorente et al., 2021). We also remove scenes with shortwave albedo less than 0.05 following de Gouw et al. (2020), which account for most of the remaining unphysical TROPOMI observations (XCH4 < 1700 ppb), and scenes north of 50°N in winter. Figure 2 shows the final 2919358 observations, regridded onto the GEOS-Chem grid and averaged seasonally. The filters preserve 69% of the high-quality retrievals and increase the GOSAT – TROPOMI correlation in all seasons, with the largest increases in winter and spring. Seasonal regional biases decrease by between 7% and 21% and are always within the one standard deviation range of both the TROPOMI and GOSAT data.

After filtering the data, we find a residual aseasonal latitudinal bias in the TROPOMI - GEOS-Chem difference. We define a latitudinal correction term (ppb) for the GEOS-Chem – TROPOMI difference using a first-order polynomial where is the degrees latitude. We also conduct sensitivity tests without the latitudinal correction, in which case we remove a mean bias of 9.11 ppb, which we attribute to errors in the boundary conditions. After all filtering and corrections, 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**

The observing system error covariance matrix (equation 1), includes contributions from the forward model, the instrument, and representation error (Brasseur and Jacob, 2017). We calculate the error 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, which we calculate on a seasonal 2° × 2° grid, 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 set a minimum error of 10 ppb, which applies to 32% observations. We find a mean observational error standard deviation of 11.5 ppb, with larger errors in winter and at high latitudes. The resulting variances are the diagonal elements of . Off-diagonal terms are assumed zero in the absence of better information, which we account for by introducing a regularization factor (Chevallier et al., 2007). We describe the choice of in section 2.7.

**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 . While Jacobian matrix construction (see Introduction) is an embarrassingly parallel problem, conducting a perturbation simulation for the 23691 grid cells optimized by this inversion is computationally intractable. We construct the Jacobian matrix at substantially decreased computational cost using the reduced-rank method introduced by Nesser et al. (2021), which is justified by the heterogeneous information content of the TROPOMI observations. 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, 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 observation grid cell *i* according to

where is a dimensionless coefficient providing a crude representation of turbulent diffusion, 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 wind speed taken here as 5 km h-1, and is the surface pressure taken here as 1000 hPa. We assume that , where gives the absolute value of the difference in latitude or longitude grid cell index between and , 36 is the sum of values, and gives the number of grid cells in the corresponding concentric ring. This representation of turbulent diffusion decreases the sparsity of , increasing its value as a first estimate. This estimate is sufficient to reproduce the dominant patterns of information content because of its the dependence on the prior error covariance matrix via the mass balance approach and on the observational density as quantified by the sparsity structure of (Nesser et al., 2021).

We use to calculate the initial patterns of information content that are perturbed in the forward model. We calculate the prior pre-conditioned Hessian (equation 2) using and complete its eigendecomposition. The resulting matrix of eigenvectors is related to the patterns of information content via , which is equivalent to the eigenvectors of the averaging kernel matrix calculated with (Bousserez and Henze, 2018). We perturb the = 434 eigenvectors that capture 50% of the DOFS generated with . We then apply an optimal operator that restores the original state dimension and minimizes information content loss to yield an updated reduced-rank Jacobian matrix estimate . We then recompute the eigenvectors, perturb the = 1952 eigenvectors that explain 80% of the initial DOFS, 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 (Houweling et al., 2014). 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 use four sets of parameters by varying the inclusion of the latitudinal correction to the model – observation difference and the use of a boundary condition correction. For each of these parameters sets, we choose the relative prior error (50%, 75%, or 100%) and regularization factor (between 0.175 and 0.5) so that the prior term of the cost function evaluated at the posterior solution averages to 1 across all grid cells optimized by the reduced-rank inversion, defined as grid cells where the averaging kernel sensitivities are greater than 0.05 (section 2.1; Lu et al., 202?). 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 parameter sets, for a total of 8 inversions. All inversions have reasonable emission ranges with low numbers of negative grid cells, most on the same order of magnitude as the soil sink. 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 by 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. 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 prior fractional sectoral contributions are correct in each grid cell and that emission sources are evenly distributed in grid cells that cross state or urban lines. The high resolution of the inversion decreases the chance of source co-location or significant distributional errors across boundaries, improving this assumption.

**3 Results and discussion**

Figure 3 shows the ensemble mean posterior correction factors relative to the prior emissions estimate (left) and ensemble mean averaging kernel sensitivities (right). Grid cells unoptimized by any inversion (mean averaging kernel sensitivity < 0.05) are shown as blank cells. Optimized grid cells explain 17% (12% - 21%) of prior emissions in Canada, 74% (65% - 83%) in the contiguous United States (CONUS), and 65% (59% - 71%) in Mexico. We find 772 (421 - 1279) DOFS across the domain, where the values in parentheses are the ensemble minimum and maximum, respectively. Of these DOFS, 37 (15 - 69) are found in Canada, 641 (350 – 1058) in CONUS, and 86 (53 - 134) in Mexico. The high fraction of optimized emissions and DOFS in CONUS reflects the co-location of emissions (figure 1) with the highest density of TROPOMI observations (figure 2). The comparatively low fraction of optimized emissions (Canada) and DOFS (Canada and Mexico) reflects lower co-location of emissions and observation and results in insignificant posterior emission changes relative to the national inventories used as prior. As a result, we focus our discussion on CONUS. We still find a large increase in information content relative to past inversions over North America: 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.

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.]

We further evaluate the reduced-rank inversion by comparing our mean posterior emissions to a full-rank regional inversion of TROPOMI observations for May 2018 to February 2020 to quantify emissions from major oil and natural gas producing regions (Shen et al., 2022). Using the same basin boundaries, we find broadly consistent posterior emissions with Shen et al. (2022), with all posterior emissions within 0.25 Tg a-1 and all but five basins within 0.10 Tg a-1. We find small but significant differences in six basins (the Delaware, the Marcellus, Eagle Ford, California, Wyoming, and the Uinta), with posterior emissions exceeding the 0.5 Tg a-1 threshold defined by Shen et al. (2022) for successful quantification of basin emissions by TROPOMI only in the Delaware. These differences could result from the different time periods examined. The broad agreement found indicates that the reduced-rank approach reproduces full-rank results.

**3.1 CONUS sectoral emissions**

We find posterior anthropogenic emissions of 30.9 (30.0 - 31.8) Tg a-1 in CONUS, an 8% (5% - 11%) increase from our prior estimate of 28.7 Tg a-1 and a 15% (12% - 19%) increase from the most recent EPA Greenhouse Gas Inventory (GHGI) estimate of 26.8 Tg a-1 for 2019. Lu et al. (2022) found larger anthropogenic methane emissions of 36.2 (32.1 – 37.6) 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) surveyed GOSAT and in situ inversions and found median posterior anthropogenic methane emissions for the United States of 26.5 (20.8 – 38.7) Tg a-1 and 31.9 (23.9 - 43.1) Tg a-1, respectively, for 2019. These large ranges encompass our result and the Lu et al. (2022) result. The difference from Lu et al. (2022) could reflect changes between 2017 and 2019, different prior emissions, or differences between GOSAT and TROPOMI, including the increased observational density provided by TROPOMI. The latter explanation is supported by the increase in DOFS we find compared to Lu et al. (2022) and by the narrower posterior emissions ranges we find in our quality-controlled ensemble compared to Lu 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. From the off-diagonal structure of (equation 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 and table 1 shows the emissions and information content data. The largest changes from the GEPA inventory and the 2022 EPA GHGI for 2019 (henceforth “GHGI”) occur in anthropogenic sectors, including landfills, livestock, oil and natural gas, and coal. For these sectors, we find sectoral averaging kernel sensitivities between 0.47 and 0.91 that are on average 61% larger than the values found by Lu et al. (2022), indicating that TROPOMI provides an improved constraint on sectoral emissions. We find little adjustment to wastewater and other anthropogenic sources due in part to their low emissions. We find a small but significant increase in wetland emissions that is consistent with the large range found by Lu et al. (2022). However, the observing system constrains a relatively small fraction of wetland grid cells.

Landfill emissions show the largest relative and absolute increase from GEPA and the GHGI for 2019. We find posterior emissions of 6.9 (6.4 - 7.5) Tg a-1, an increase of 21% (12% - 32%) relative to GEPA and 52% (41% - 65%) relative to the GHGI. The larger discrepancy with respect to the GHGI results from an assumed downward trend in landfill emissions since 1990 due to increased landfill gas collection and decreased organic material in landfills. Lu et al. (2022) found similar mean posterior landfill emissions of 7.5 (5.9 – 7.7) Tg a-1 for 2017. 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 3.4.

Livestock emissions show the second largest absolute increase from the prior emissions estimate, with posterior emissions of 10.4 (10.0 - 10.7) Tg a-1 representing an increase of 13% (9% - 16%) from GEPA and 11% (6% - 14%) from the GHGI. The smaller discrepancy with respect to the GHGI reflects the increase in CONUS cattle populations since 1990. Lu et al. (2022) found similar mean posterior livestock emissions of 10.4 (8.8 - 11.6) Tg a-1 over CONUS for 2017. 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 with our posterior livestock estimate of 5.4 (5.1 - 5.6) Tg a-1 over the same region. We attribute the low GHGI value to underestimated manure management emissions, which we discuss in section 3.2.

TROPOMI provides the strongest constraint for fossil fuel emissions with sectoral averaging kernel sensitivities of 0.91 for oil and natural gas and 0.6 for coal. Posterior oil and natural gas emissions are 10.4 (10.1 - 10.7) Tg a-1, an increase of 11% (7% - 14%) from the GEPA inventory updated for 2018 combined with the high-resolution EDF inventory over the Permian and 18% (14% - 22%) from the GHGI. The larger discrepancy from the GHGI reflects decreased production emissions since 2018. Lu et al. (2022) found much larger posterior emissions of 4.8 (3.1 - 4.9) Tg a-1 for oil and 8.9 (8.0 - 9.8) Tg a-1 for natural gas. However, they found decreasing natural gas emissions beginning in 2014. Compared to Lu et al. (2022), Shen et al. (2022) found lower oil and natural gas emissions of 12.6 ± 2.1 Tg a-1 from an inversion of TROPOMI data over 14 basins extrapolated to the national scale for May 2018 to 2020, which is consistent with continued emissions decreases into 2019. Coal mining emissions exhibit the largest decline in sectoral emissions, decreasing 48% (34% - 59%) from GEPA and 30% (11% - 44%) from the GHGI. Lu et al. (2022) found much larger posterior emissions of 2.9 (2.3 - 3.4) Tg a-1 for 2017, but our decrease is consistent the 30% decrease in CONUS coal production since 2012 (USGS). Deng et al. (2022) found cumulative fossil fuel emissions of 9.8 (8.1 - 13.7) Tg a-1 from an ensemble of GOSAT inversions and 12.6 (8.0 - 16.7) Tg a-1 for an ensemble of in situ inversions in 2019, both of which are consistent with our result.

**3.2 Livestock emissions**

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

**3.3 State emissions**

We partition emissions, excluding offshore emissions, to each of the 48 states in CONUS as described in section 2.8 and compare the results to GEPA, the EPA’s new state inventory, which is scaled to match the GHGI (henceforth “state GHGI”), and inventories prepared independently by state governments. Figure 5 shows state prior and posterior emissions for the 29 states responsible for 90% of posterior CONUS anthropogenic emissions, excluding offshore emissions, and table S1 shows the full results. TROPOMI provides a strong constraint at this resolution, with sensitivity to TROPOMI observations 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.

We find a large increase in state emissions compared with EPA estimates, with state posterior emissions on average 6% larger than GEPA and 12% larger than the state GHGI. The bigge

st increases from the state GHGI occur in the top 10 methane-producing states, where state posteriors are on average 24% larger than GEPA and 38% larger than the state GHGI. These states are responsible for 55% of posterior CONUS methane emissions, compared to 50% of GEPA emissions and 47% of state GHGI emissions. We find a significant emissions increase compared to the state GHGI in eight of these ten states. These states are dominated by oil and natural gas (Texas, New Mexico, Louisiana, and Oklahoma), livestock (California and Iowa), and landfills (Florida and Illinois). Oil and natural gas emissions have a disproportionate influence, generating 47% of mean posterior emissions and 45% of the observed increase, compared to the national averages of 32% and 36%, respectively. Consistent with our sectoral analysis, the largest absolute negative posterior - state GHGI differences are found in historically coal-producing states, including Wyoming, West Virginia, and Pennsylvania.

We consider in more detail Texas and California, which are responsible for 27% of posterior emissions. Texas alone produces 6.3 (6.1 - 6.5) Tg a-1, 21% of posterior emissions (18% in GEPA) and three times more than California. These emissions are a 22% increase from GEPA and a 69% increase from the state GHGI. We also find a different sectoral partitioning compared to the state GHGI. Oil and natural gas emissions account for 69% of posterior emissions compared to 56% in the state GHGI, with 46% of all posterior emissions occurring in the Permian basin, the largest oil and natural gas producing region in CONUS (Zhang et al., 2020). We find Permian emissions of 2.9 Tg a-1 that are consistent with recent studies (Zhang et al., 2020; Schneising et al., 2020; Liu et al., 2021; Shen et al., 2022; Varon et al., in review).

California produces 2.1 (2.0 - 2.1) Tg a-1. We find smaller emissions than the 2.86 ± 0.21 Tg a-1 found by Wecht et al. (2014) in an inversion of observations from the 2010 (May – June) CalNex aircraft campaign, but this could be due to decreased emissions or seasonal differences. Our posterior emissions are a 16% increase from GEPA, a 33% increase from the state GHGI, and a 32% increase from the independent estimate prepared by the California Air Resource Board (CARB). Relative to the CARB inventory, 78% of the inferred increase is attributable to livestock and 14% to oil and natural gas. We find good agreement with the sectoral partitioning in both the state GHGI and the CARB inventory. Livestock explain 54% of emissions in the posterior, 50% in the state GHGI, and 54% in the CARB inventory, while landfills explain 25%, 23%, and 21% of emissions, respectively. We find slightly smaller relative contributions from oil and natural gas, which is responsible for 11% of posterior emissions compared to 20% and 17% of state GHGI and the CARB inventory respectively. This partitioning differs from that found in Wecht et al. (2014), where 30% of emissions were attributed to livestock, 38% to landfills, and 22% to oil and natural gas. As above, the discrepancy could be due to temporal or seasonal differences, or from the coarser resolution (1/2° × 2/3°) of the CalNex inversion.

We also compare our posterior emissions to other available state greenhouse gas inventories for the year closest to 2019 where the observing system provides a strong constraint (state averaging kernel sensitivity greater than 0.5). Of the inventories of Colorado, Iowa, Louisiana, and Pennsylvania, our posterior agrees only with Pennsylvania. However, 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 Iowa, and Louisiana are smaller than our posterior emissions. In Iowa, our prior estimate is only 4% smaller than the inventory value, but we find a 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 65% increase in emissions attributed to oil and natural gas (71%) and other sources, including rice (16%). The Colorado inventory is larger than our prior and posterior estimates by 44% and 65%, respectively, with oil and natural gas emissions more than twice as large as our prior.

**3.4 Urban area emissions**

We calculate the posterior emissions for 219 urban areas across CONUS, providing the first comprehensive analysis of urban emissions in CONUS. Urban areas are defined using the U.S. Census Urban Areas for 2010, limited to cities with populations greater than 1e6 according to the 2010 Census and with urban area averaging kernel sensitivities greater than 0.05. With a mean urban area averaging kernel sensitivity of 0.23 (0.11 - 0.34), the observing system provides a weaker constraint on urban areas than on sectors or states due to smoothing errors, larger instrument errors incurred by the heterogeneous urban grid, and increased model transport errors at high resolution. We still find significant corrections: urban anthropogenic posterior emissions are 2.4 (2.2 - 2.7) Tg a-1, 29% (16% - 44%) larger than GEPA. These emissions represent 8.3% (7.7% - 7.2%) of anthropogenic emissions, equivalent to almost a quarter of CONUS oil and natural gas emissions. The top 10 methane-producing cities, shown in figure 6 and described in table S2, have a larger mean urban averaging kernel sensitivity of 0.40 and are responsible for 40% (39% - 42%) of urban posterior emissions. Boston is excluded due to the limited constraint at high latitudes, resulting in smaller corrections and lower emissions. We find a mean increase relative to GEPA of 69% in these cities and emissions more than twice as large as the inventories from New York City and Philadelphia, the only available methane emission estimates among these cities.

We compare our results to 17 studies of ten urban areas published since 2015. These studies tended to quantify emissions in coastal metropolitan centers, with only three studies quantifying emissions in smaller, non-coastal cities: Kansas City (Plant et al., 2022) and Indianapolis (Lamb et al., 2016 and Jones et al., 2021). On average, we find lower emissions than past studies, which may result in part from the weaker constraint generated by TROPOMI and in part from from our restrictive definition of urban extent. We find the largest discrepancies over coastal cities, including San Francisco (Fairley and Fischer, 2015; Jeong et al., 2016, 2017; Guha et al., 2022), Washington (D.C.) and Baltimore (Huang et al., 2016; Ren et al., 2018; Lopez-Coto et al., 2020; Plant et al., 2019, 2022), Boston (Plant et al., 2019, 2022; Sargent et al., 2021), and Los Angeles (Cui et al., 2015; Jeong et al., 2016; Wunch et al., 2016; Yadav et al., 2019; Cusworth et al., 2022). In Philadelphia (Plant et al., 2019, 2022) and Atlanta (Plant et al., 2022), emissions estimated using observed methane-carbon monoxide ratios together with carbon monoxide inventories were consistent with our posterior. However, the confidence intervals in both studies spanned more than 200 Gg a-1, and Plant et al. (2019) found Philadelphia emissions significantly larger than our estimate using methane-carbon dioxide ratios. We find consistent results with most surveyed studies in New York City and Indianapolis (Plant et al., 2019, 2022; Pitt et al., 2022; Lamb et al., 2016; Jones et al., 2021).

While the observed increase in urban emissions is likely an underestimate, it still suggests that landfill emissions or natural gas distribution or post-meter emissions are too low. The EPA added post-meter emissions of 456 Gg a-1 to the GHGI in 2022. This addition explains 82% (57% - 130%) of the increase from GEPA in urban areas. We are unable to attribute the remaining discrepancy to a particular sector due to source co-location. We also find no correlations between the posterior increase from GEPA and 2010 urban area population, population change from 2000 to 2010, population density, or surface area. Indeed, the three largest cities by population (New York, Los Angeles, and Chicago) have the smallest per capita methane emissions among the top 10 methane-producing cities, while the two smallest cities by population (Detroit and Indianapolis) have the largest per capita emissions. The lack of correlation reflects the variability of methane emission sources and trends between urban areas. Floerchinger et al. (2021) found “dramatically different” emission profiles in an aircraft survey of methane and ethane over seven U.S. cities, with the fraction of emissions attributable to natural gas ranging from 0.32 to 1.0. Sargent et al. (2021) surveyed 12 studies spanning six cities and found that natural gas contributions to urban emissions ranged from 43% to 88%. Both studies included geographically diverse cities with a range of populations. Plant et al. (2019) used aircraft data over six cities on the East Coast of CONUS and found that on average between 80% and 110% of emissions in each city were explained by natural gas emissions. However, their confidence intervals ranged from 45% to 170% of urban emissions.

**4 Conclusions**

[Insert.]