**High-resolution U.S. methane emissions inferred from an inversion of 2019 TROPOMI satellite data: contributions from individual states, urban areas, and landfills**

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We quantify 2019 methane emissions in the contiguous U.S. (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) Greenhouse Gases Inventory (GHGI) serves as the prior estimate for the inversion. We optimize emissions and quantify observing system information content through analytical minimization of a Bayesian cost function with eight ensemble members. We achieve high resolution with a reduced-rank characterization of the observing system to approximate the Jacobian matrix in a manner that optimally preserves information content. Our optimal (posterior) estimate of anthropogenic emissions in CONUS is 30.9 (30.0 - 31.8) Tg a-1, where the values in parentheses give the spread of the 8-member inversion ensemble. This is a 16% increase from the most recent EPA GHGI estimate of 26.7 Tg a-1 in 2019. We partition emissions by sector, with livestock emissions of 10.4 (10.0 - 10.7) Tg a-1, oil and gas of 10.4 (10.1 - 10.7) Tg a-1, coal of 1.5 (1.2 - 1.9) Tg a-1, landfills of 6.9 (6.4 - 7.5) Tg a-1, wastewater of 0.6 (0.5 - 0.7), and other anthropogenic sources of 1.1 (1.0 - 1.2) Tg a-1. Relative to the GHGI, we find the largest increase (53%) for landfills and a 29% decrease for coal. Comparison with bottom-up emission inventories from 73 individual landfills reported by the Greenhouse Gases Reporting Program (GHGRP) finds large difference that we attribute to excessive emission decay rates in the inventories, overestimated gas recovery efficiencies, and under-accounting of leaks. We exploit the high resolution of our inversion to quantify emissions in the 48 individual states in CONUS and compare to EPA’s new state-level inventories as well as to independent inventories produced by state agencies. Our posterior emissions are on average 38% larger than the EPA GHGI inventories in the largest 10 methane-producing states, with the largest upward adjustments in states with large oil and gas emissions and the largest downward adjustments in states with large contributions from coal emissions. We also calculate emissions for 95 geographically diverse urban areas in CONUS. Emissions for these urban areas are 42% larger than the GHGI, with a 66% increase in the largest 10 methane-producing cities. We attribute the discrepancy to underestimated landfill and gas distribution emissions.

**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 their anthropogenic methane emissions including contributions from individual sectors including oil and gas, coal, livestock, rice agriculture, landfills, and wastewater. 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 observations of atmospheric methane together with an atmospheric transport model to infer emissions through inverse analyses. These top-down emission estimates are most useful if they achieve high spatial resolution and maximize the information content of the observation-model system. Here we use column methane observations from the TROPOMI satellite instrument in a reduced-rank analytical inversion to infer methane emissions and the associated information content at 0.25° × 0.3125° (≈25 × 25 km2) resolution over the contiguous U.S. (CONUS) 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 used extensively in inverse analyses of methane emissions (Streets et al., 2013; Jacob et al., 2022). Past satellite instruments were limited by large pixel sizes (SCIAMCHY, 2003 – 2012) or sparse observations (GOSAT, 2009 – present). The Tropospheric Monitoring Instrument (TROPOMI) launched in 2017 aboard the Sentinel-5 Precursor satellite 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), though with a success rate of only 3% limited by cloud cover, optically dark surfaces, and heterogeneous terrain (Hasekamp et al., 2019). Inversions of TROPOMI data allow for high-resolution quantification of methane emissions but require understanding of 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). The emissions represent the state vector for the inversion, and the CTM represents the forward model. Optimization 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, as in the case of methane, the optimal (posterior) solution and the associated errors and information content can be found analytically (Brasseur and Jacob, 2017). However, this requires the computationally expensive construction of the Jacobian matrix that represents the relationship between emissions and concentrations in the CTM. This matrix is typically constructed by conducting a CTM perturbation simulation for each emission element, limiting either the spatial resolution of the optimized emissions or the size of the inversion domain. In Nesser et al. (2021), we demonstrated an alternative method that approximates the Jacobian matrix by perturbing emission patterns that are informed by both the prior emissions and the observations. This approach optimally exploits the information content of the observations, quantifying emissions at the highest resolution possible where the observing system provides a constraint and defaulting to the prior estimate elsewhere.

Many inverse studies that quantified U.S. methane emissions using surface, aircraft, or satellite observations have found large discrepancies with the U.S. Environmental Protection Agency’s Greenhouse Gas Inventory (GHGI), the bottom-up emissions estimate reported to the UNFCCC. Wecht et al. (2014) found livestock emissions 40% larger than the GHGI for the summer of 2004. Miller et al. (2013) inferred emissions 50% larger than the GHGI for 2007 and 2008, which they attributed to underestimated oil, gas, and livestock emissions. Turner et al. (2015) found similar results for emissions from 2009 to 2011 compared to the GHGI. Maasakkers et al. (2021) inferred oil and gas emissions 35% and 22% higher than the GHGI, respectively, for 2010 to 2015. Lu et al. (2022) found mean 2010 – 2017 anthropogenic emissions 10.0 (6.5 - 11.8) Tg a-1 larger than the GHGI, which they attributed largely to oil emissions.

Higher resolution regional studies have targeted specific aspects of U.S. emissions, including emissions from different sectors, states, and urban areas. Karion et al. (2015) found oil and gas emissions in the Barnett Shale of eastern Texas consistent with the GHGI but larger than e reported by industry to the EPA’s Greenhouse Gas Reporting Program (GHGRP). A series of studies have inferred much higher emissions in the Permian Basin than implied by the GHGI (Zhang et al., 2020; Schneising et al., 2020; Liu et al., 2021; Shen et al., 2022; Chen et al., 2022; Varon et al., in review). Chen et al. (2018) and Yu et al. (2021) found underestimated livestock emissions in the upper Midwest. Jeong et al. (2016) inferred California emissions 20% to 80% larger than a state inventory from the California Air Resources Board (CARB). Plant et al. (2019) found methane emissions from six East Coast urban areas to be more than two times larger than the GHGI.

Here we use the reduced-rank method of Nesser et al. (2021) in an analytical inversion of 2019 TROPOMI observations at 0.25° × 0.3125° resolution over North America (CONUS, Canada, and Mexico) using national emission inventories reported to the UNFCCC as prior estimates. Several filters are applied to the TROPOMI data to avoid biases. We focus our analysis on CONUS, with particular attention to emissions from individual landfills, states, and urban areas. We compare our results to the most recent version of the GHGI (EPA, 2022). This inventory includes for the first time emissions estimates for individual states, for which our inversion provides the first observational evaluation. We also compare our results to inventories prepared by individual states and cities.

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

We conduct an ensemble of inversions of 2019 TROPOMI methane observations over the North American domain shown in Figure 1 (9.75°N - 60°N, 130°W - 60°W) using the nested GEOS-Chem CTM at 0.25ox0.3125o horizontal resolution over that domain as forward model. The TROPOMI observations are fit to simulated GEOS-Chem concentrations to optimize mean methane emissions for 2019 at the native 0.25° × 0.3125° resolution of GEOS-Chem. This corresponds to state vector with dimension = 23691 including all grid cells with prior methane emissions larger than 0.1 Mg km-2 a-1, accounting for over 99% of North American methane emissions. In a subset of the ensemble, we optimize boundary conditions corresponding to each of the four cardinal directions (north, south, east, and west) for the nested GEOS-Chem simulation. Methane chemical and soil sinks are not optimized because they are relatively uniform and slow compared to the ventilation timescale of the domain.

**2.1 Reduced-rank analytical inversion**

The inversion uses observed concentrations arranged in a vector to optimize gridded emissions arranged in the state vector by minimizing a Bayesian cost function regularized by the prior emissions estimate and assuming normal errors:

The prior and observing system error covariance matrices and , respectively, are assumed diagonal in the absence of better information. The regularization factor corrects for the absence of covariance in (Chevallier et al., 2014). We generate an inversion ensemble using a range of prior error variances and values to capture the inversion’s sensitivity to uncertainty in these parameters (section 2.7). The reduced-rank Jacobian matrix represents the sensitivity of concentrations to emissions in the CTM (Rodgers, 2000). To construct a rank Jacobian matrix for the 0.25° × 0.3125° GEOS-Chem grid at reduced computational cost, we perturb in the CTM the emission patterns that best capture the prior emissions and the information content of the TROPOMI observations as described in section 2.6.

Analytical minimization of the cost function following Rodgers (2000) 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. However, this solution requires inverting an matrix, which produces numerical instabilities due to the rank reduction in the Jacobian construction. Here we use a reduced-rank approximation of the posterior solution following Bousserez and Henze (2018) to solve the inversion on an orthonormal basis that optimally spans the information content of the satellite–forward model observing system. The basis is given by the eigendecomposition of the prior-preconditioned Hessian of the cost function

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 , which we address with Dask, a Python parallelization package. The reduced-rank posterior approximation is then generated using the largest eigenvalues and the associated eigenvectors (Bousserez and Henze, 2018):

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 the reduced-rank Jacobian matrix. We choose to maximize the DOFS within the available computational resources (section 2.8). The diagonal elements of are often referred to as averaging kernel sensitivities and their sum (trace of ) gives the degrees of freedom for signal (DOFS) that represent the number of pieces of information independently quantified by the observing system (Rodgers, 2000; Bousserez and Henze, 2018). The reduced-rank inversion and Jacobian matrix do not attempt to optimize emissions in areas with low information content, so we default to the prior estimate for grid cells with averaging kernel sensitivities less than 0.05 (Nesser et al., 2021).

**2.2 Prior estimates and errors**

Figure 1 shows the prior emission estimates for different sectors. Anthropogenic emissions are from the spatially disaggregated (gridded) versions of the EPA GHGI for the U.S. for 2012 (Maasakkers et al., 2016), the INECC inventory for Mexico for 2015 (Scarpelli et al., 2020), and the ECCC inventory for Canada for 2018 (Scarpelli et al., 2021). We update GHGI oil and gas emissions to 2018 following Shen et al. (2022) and use the Environmental Defense Fund’s inventory for the Permian basin for 2019 (Y. Zhang et al., 2020). We treat oil and 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 from 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 monthly scaling factors as described by Maasakkers et al. (2016) and Zhang et al. (2016), respectively.

Prior emissions for wetlands are given by the high-performance subset of 9 members of the WetCHARTs ensemble version 1.3.1 that best match global GOSAT inversion results (Ma et al. 2021). Lu et al. (2022) found in an inversion of GOSAT data over North America that the high-performance subset overestimated wetland methane emissions, particularly at high latitudes. We remove from the ensemble the two members (models 1923 and 2913) that are most responsible for this overestimate. Other natural methane emission sources are minor and include open fires, termites, and geological seeps, for which we follow the emissions described in Lu et al. (2022).

We assume uniform relative error standard deviations for prior emissions of between 50% and 100% for the different members of our inversion ensemble. Previous inversions that optimized methane emissions over North America assumed 50% error standard deviation on prior emissions (Maasakkers et al., 2021; Lu et al., 2022). We inflate errors up to 100% in our ensemble to account for increased error at high resolution (Maasakkers et al. 2016).

**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 sinks from OH, Cl, soil uptake, and stratospheric oxidation are as described in Maasakkers et al. (2019). Initial conditions for January 1, 2019 and 3-hourly boundary conditions 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).

**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 in the 2.3 μm methane absorption band 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 is only 3% successful due to cloud cover, variable topography, low or heterogeneous albedo, or high aerosol loading (citation). We use retrieval v14 as 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.

Previous analyses of TROPOMI data identified surface artifacts (Barré et al., 2021) and spatially variable biases relative to the more accurate but sparser GOSAT data (Qu et al., 2021; Chen et al., 2023). We filter the data to remove snow- and ice-covered scenes using blended albedo, an empirical parameter developed by Wunch et al. (2011) and suggested for the TROPOMI data by Lorente et al. (2021). We remove scenes with blended albedo > 0.75 in non-summer seasons. We also remove scenes with albedo in the shortwave infrared 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 used for the inversion on the GEOS-Chem 0.25° × 0.3125° grid, and Figure S1 shows the improved correlation with seasonal averaged GOSAT observations on a 2° × 2° grid. The filters preserve 69% of the high-quality retrievals of TROPOMI v14 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. Comparison to a GEOS-Chem simulation driven by the prior emissions shows a mean GEOS-Chem – TROPOMI bias of = 9.1 ppb over North America. This bias can be fit as a linear function of degrees latitude as . It could be caused by errors in the boundary conditions. We correct it in our inversion ensemble members by removing either the continental mean bias or a latitude-dependent correction from the GEOS-Chem concentrations.

**2.5 Observing system errors**

The observing system error covariance matrix , includes contributions from the forward model, instrument, and representation errors (Brasseur and Jacob, 2017). We calculate the total observing system 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 defines the standard deviation of the observing system. We set a minimum error standard deviation of 10 ppb, which applies to 32% observations. We find a mean observing system error standard deviation of 11.5 ppb, with the largest 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 the regularization factor (Chevallier et al., 2007). We describe the choice of in section 2.7.

**2.6 Jacobian matrix**

Constructing the Jacobian matrix for our inversion would typically require conducting a 1-year perturbation simulation for each of the = 23691 grid cells optimized. This 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 takes advantage of 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 described 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 the gravitational acceleration, is the wind speed taken here as 5 km h-1, and is the surface pressure taken here as 1000 hPa. The use of produces off-diagonal structure in , which we found in Nesser et al. (2021) to be necessary for an effective first estimate. We apply a simple isotropic turbulent diffusion scheme in which the influence of emissions spreads linearly to concentric rings of grid cells. This is represented as , where gives the absolute value of the difference in latitude or longitude grid cell index between and , 36 is the sum of values, gives the number of grid cells in the corresponding concentric ring, and .

We use together with the error covariance matrices and 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 perform 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

**2.7 Inversion ensemble**

The posterior error covariance matrix that results from Bayesian optimization in equation (4) does not account for errors in inversion parameters (Houweling et al., 2014). The analytical solution readily allows for the creation of an ensemble of inversions that reflects the sensitivity of the results to the chosen setup including paraeters. Table 1 summarizes our ensemble of inversions. We conduct inversions that do and do not optimize the boundary conditions, and that do and do not apply a latitudinal correction to the prior (model – observation) difference. For each inversion, 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, as expected from the narrow chi-square distribution (Lu et al., 2021). This yields an ensemble of 8 quality-controlled inversions with indistinguishable validity. We report the mean posterior emissions for the ensemble, 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 individual source sectors or regions, including states, and urban areas. We aggregate the native resolution emissions and errors estimates to the corresponding sectors, states, or urban areas using a summation matrix . The rows of are given by the relative contribution of each grid cell to each individual source category. For sectoral attribution, the rows are given by the relative contribution of each grid cell to a given sector in the prior emissions estimate. For state attribution, the rows are given by the fraction of each grid cell within a given state. For urban area attribution, the rows have binary values depending on whether or not the grid cell overlaps with a given urban area. The reduced-dimension posterior estimate, posterior covariance matrix, and averaging kernel matrix 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 lines. The high resolution of the inversion decreases the chance of source co-location or significant distributional errors across boundaries.

**3 Results and discussion**

Figure 3 shows the ensemble mean posterior scale factors relative to the prior emissions estimate (left) and the corresponding averaging kernel sensitivities (right). Grid cells unoptimized by the inversion (mean averaging kernel sensitivity < 0.05) are left blank. We find 772 (421 - 1279) DOFS across the domain, where the values in parentheses are the ensemble minimum and maximum, respectively. This represents a large increase in information content relative to the previous North America inversion of Lu et al. (2022), who found 114 DOFS in a joint inversion of GOSAT and NOAA’s GLOBALVIEWplus ObsPack in situ data. This increase reflects both the improved constraint provided by TROPOMI and the benefit of achieving 0.25° × 0.3125° resolution on the continental scale. 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 DOFS in CONUS reflects both the large emissions (Figure 1) and the high density of TROPOMI observations (Figure 2). As a result, we focus our discussion on CONUS, where we isolate anthropogenic emissions by removing contributions from wetlands and other natural sources following Section 2.8. We compare our posterior emissions the most recent EPA GHGI inventory for 2019, which includes emissions estimates for individual states (EPA, 2022).

Figure 4 shows simulated observations from GEOS-Chem driven by the prior and mean posterior emissions compared to TROPOMI data and to independent in situ surface and aircraft observations from NOAA’s GLOBALVIEWplus CH4 ObsPack v3.0 database (cite). We follow Lu et al. (2021) and use only daytime average Obspack surface and tower observations with outliers excluded. The posterior GEOS-Chem simulation decreases the root mean squared error (RMSE) of the simulated observations with the TROPOMI data relative to the prior simulation by 5% (from 15.8 ppb to 15.0 ppb) and the mean bias by 9% (from 9.1 ppb to 8.3 ppb). Both simulations produce similar Pearson correlation coefficients. When compared to ObsPack data, both simulations yield similar RMSE, mean bias, and R2, while the posterior simulation improves the reduced major axis regression (RMA) intercept by 4% and the slope by 3%. The broad agreement of both simulations with observations reflects the high quality of the prior emissions estimate in North America (Maasakkers et al., 2019). The failure of the posterior simulation to significantly improve agreement with the observations compared to the prior simulation may reflect the large observing system errors for individual observations.

**3.1 CONUS sectoral emissions**

We find posterior anthropogenic methane emissions of 30.9 (30.0 - 31.8) Tg a-1 in CONUS, a 16 (12 - 19) % increase from the most recent GHGI estimate for 2019 (henceforth “GHGI”) of 26.7 Tg a-1 (EPA, 2022). Lu et al. (2022) found larger anthropogenic 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) reviewed an ensemble of global inversions and found median U.S. posterior anthropogenic emissions for 2019 of 26.5 (20.8 - 38.7) Tg a-1 with GOSAT data and 31.9 (23.9 - 43.1) Tg a-1 with in situ data.

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), indicating that we can separate sectoral emissions. Figure 5 and Table 2 summarize the results. Livestock, oil and gas, and landfills account for 90% of posterior anthropogenic emissions and all increase relative to the GHGI. We find a significant decrease from the GHGI only for coal. For these four sectors, we find sectoral averaging kernel sensitivities between 0.47 and 0.91, significantly larger than the values found by Lu et al. (2022) from GOSAT and in situ data. 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 only constrains about half of wetland emissions, with most of the inferred increase limited to the southeastern coast, including in South Carolina, Georgia, and eastern Florida.

Landfill emissions show the largest relative and absolute increase from the GHGI for 2019. We find posterior emissions of 6.9 (6.4 - 7.5) Tg a-1, an increase of 52% relative to the GHGI estimate of 4.5 (3.5 – 5.5) Tg a-1, where the values in parentheses represent EPA bottom-up error estimates (95% confidence interval). 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 EPA underestimate to two components of landfill inventory methodologies. First, for landfills with gas recovery systems, the GHGI relies on reported emissions from the GHGRP that use too-high collection efficiencies. Second, the GHGI does not account for site-specific operations that may produce anomalous emissions. We discuss these causes in detail in Section 3.2.

Coal mining emissions of 1.5 (1.2 - 1.9) Tg a-1 exhibit the largest decrease in sectoral emissions relative to the GHGI estimate of 2.1 (2.0 - 2.5) Tg a-1. Lu et al. (2022) found much larger posterior emissions of 2.9 (2.3 - 3.4) Tg a-1 for 2017 that are consistent with GHGI estimates for 2012. Our lower estimate better reflects the 30% decrease in CONUS coal production from 2012 to 2019 (EIA, 202x), which is also shown in the 30% decrease in GHGI coal emissions over the same period. As expected, emissions correlate with underground coal mining: Appalachia generates 56% of coal from underground mines and 64% of posterior emissions, while the Illinois Basin yields 30% of underground coal and 20% of posterior emissions.

Livestock emissions show broad agreement with the GHGI, with posterior emissions of 10.4 (10.0 - 10.7) Tg a-1 representing an insignificant 11% increase from the GHGI estimate of 9.4 (8.6 - 10.2) Tg a-1. 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 U.S. 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 (5.2 - 5.6) Tg a-1 over the same region. Despite agreement with total GHGI livestock estimates, we find a significant increase in manure management emissions from 2.3 Tg a-1 to 3.1 Tg a-1, which would almost entirely explain the observed discrepancy between the mean GHGI and posterior emissions. The increase in manure management emissions is concentrated over the California Central Valley, northern Iowa, and Sampson and Duplin Counties in North Carolina. Notably, Iowa is the largest pork-producing state, and Sampson and Duplin Counties are the two largest pork-producing counties in CONUS (USDA, 2017). We find no correlation between our inferred increase and hog populations, which could reflect variability in manure management practices.

Posterior oil and gas emissions are 10.4 (10.1 - 10.7) Tg a-1, an 18% increase from the GHGI estimate of 8.7 (7.4 – 10.2) Tg a-1. 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 gas in 2017. However, they found decreasing gas emissions beginning in 2014. Compared to Lu et al. (2022), Shen et al. (2022) found lower oil and gas emissions of 12.6 ± 2.1 Tg a-1 from an inversion of TROPOMI data over 14 North American basins extrapolated to the national scale for May 2018 to 2020, which is consistent with continued emissions decreases into 2019. These emissions are within the uncertainty range of our posterior estimate, and we find consistent basin-level results as shown in Figure S2. All posterior basin emissions are within 0.25 Tg a-1 of Shen et al. (2022) and all but five basins are within 0.10 Tg a-1. Of the basins where posterior emissions exceed the 0.5 Tg a-1 threshold defined by Shen et al. (2022) for successful quantification of basin emissions by TROPOMI, we find significant but small differences only in the Delaware. We find total emissions in the Permian basin of 2.9 Tg a-1, which is consistent with other recent studies (Zhang et al., 2020; Schneising et al., 2020; Liu et al., 2021; Varon et al., 2023).

**3.2 Landfill emissions**

We consider in more detail the 52% increase in our posterior landfill emissions relative to the GHGI. The GHGI uses two methodologies to estimate landfill emissions (EPA, 2022). The first uses a first order decay model based on landfill mass so that emissions peak the year after waste disposal (IPCC, 2006). Recovered methane is removed from the total. However, a survey of 128 California landfills with gas recovery systems found that methane was produced at relatively constant rates over time (Spokas et al., 2015). The second method back-calculates emissions as a function of recovered methane given an estimated collection efficiency based on cover and operation methods. A default efficiency of 0.75 is assumed if cover information is unavailable (40 CFR § 98.343). GHGI estimates also incorporate GHGRP information from 1297 landfills emitting more than 1 Gg a-1 across the U.S., 500 of which have gas recovery systems (EPA LMOP, 2022). GHGRP methane emissions can be estimated with two of four methods depending on whether gas is collected (EPA, 2022; 40 CFR § 98.343). Landfills without gas collection estimate emissions based on landfill attributes and the GHGI first-order decay model. Landfills with gas collection must use one of these methods in addition to a back-calculation approach. These methods have high uncertainties and have not been field validated (NAS, 2018).

We compare our posterior landfill emissions to GHGRP facilities reporting more than 2.5 Gg a-1 methane. We limit our analysis to grid cells where TROPOMI provides an averaging kernel sensitivity greater than 0.20 and where landfills explain more than 50% of prior grid cell (0.25o×0.3125o) emissions so that we are confident of our ability to separate landfill emissions from other sources. We exclude 33 facilities in grid cells containing multiple landfills because we are unable to separate the individual contributions to total emissions. Figure 6 shows the posterior emissions and corrections to GHGRP for the remaining 73 facilities and Table 3 shows GHGRP and posterior information for the top 10 methane-producing landfills as ranked by posterior emissions.

We validate our posterior landfill results by comparison to aircraft-derived estimates for nine facilities as shown in Figure 6. We find agreement within error bounds at the Seneca Meadows landfill in New York (landfill c in Figure 6) and at the Kiefer (d), Frank R. Bowerman (f), Altamont (g), Newby Island (h), and Keller Canyon (i) landfills in California. We find much larger emissions than previous studies at the South Side landfill (a) in Indiana and at the West Miramar Sanitary (b) and Puente Hills (e) landfills in California. The South Side landfill began construction on a large landfill gas facility in June 2019, emissions from which were not captured by the 2011 aircraft campaign. The West Miramar Sanitary landfill leaked methane at 8662 ppm in November 2019 (San Diego Air Pollution Control District, 2019), a period that was not covered by either study of the facility. The Puente Hill Landfill closed in 2013 but was previously one of the largest landfills in CONUS (cite). As a result, our prior estimate from 2012 may misattribute emissions to the landfill compared to other sources.

We find larger mean facility emissions of 13 Gg a-1 compared to the GHGRP mean of 7.2 Gg a-1 for the 73 landfills considered here. We find no correlation (R2 = 0.00) between GHGRP emissions and our posterior estimates, which does not improve when we consider only facilities that do or do not capture landfill gas. This is why there is no relationship between GHRG-reported emissions and our posterior estimates in Table 3. The implication is that the bottom-up approaches have little predictability.

For the 38 facilities that recover gas, we use reported avoided methane emissions together with posterior and GHGRP emissions to calculate a posterior and reported recovery efficiency, respectively (LMOP, 2019). We find a low correlation (R2 = 0.17) between the efficiencies regardless of facility size. The average posterior recovery efficiency of 0.50 (0.33 - 0.54) is much smaller than the GHGRP mean of 0.61, and both are much smaller than the 0.75 default (40 CFR § 98.343). The discrepancy between posterior and GHGRP efficiencies does not depend on facility age or size. Across the six landfill gas facilities at the top 10 methane-producing landfills, we find a mean posterior recovery efficiency of 0.33 that is half the GHGRP value of 0.65. Indeed, four of the six facilities report methane emissions and recovery values that are consistent with efficiencies larger than the 0.75 default. We find a similar but still lower efficiency at the Seminole Road MSW Landfill (landfill 8) and a marginally higher recovery efficiency only at Sampson County Disposal, LLC (10).

We consider in detail the 34 facilities for which posterior emissions show a significant 50% difference from the GHGRP. We find larger emissions for 29 of these facilities, with the largest discrepancies occurring in nine of the top 10 methane-producing landfills. The facilities with the largest posterior - GHGRP differences experienced significant operational changes in the last decade. National Serv-All Landfill (landfill 1) began receiving additional waste after a nearby landfill closed in March 2019 (cite). The South Shelby (2) and South Side (3) Landfills started construction on large landfill gas facilities in 2019 (cite). Finally, the City of Dothan Sanitary Landfill (6) has been full since 2014 (citation). Reported emissions peaked at 7.4 Gg a-1 that year, a value almost five times smaller than our posterior emissions, suggesting that the first order decay model is inadequate to reproduce methane emissions over time. We also find a history of air quality and landfill standard violations at these 34 facilities. At the West Miramar Sanitary Landfill (b), emissions of 8662 ppm were recorded in November 2019 (San Diego Air Pollution Control District, 2019). The Sussex County Landfill in Virginia was fined USD 99000 in 2016 for failing to address cracks in the landfill cover (citation), and we find emissions 2.3 (1.6 - 3.4) times larger than reported. Lastly, the Newby Island Landfill (h), received 30 violation notices from 2014 to 2020, including for gas leaks (citation).

There are five facilities for which our posterior emissions are significantly smaller than the 2019 GHGRP by 50%. Three report in fact large decreases in estimated methane emissions from 2019 to 2020 that imply changes in methodology. The updated estimates are statistically consistent with our posterior emissions in two cases and within 30% of our posterior emissions in the third case

**3.3 State emissions**

The EPA recently began to disaggregate the GHGI by state (EPA, 2022). The EPA uses the same methods to partition emissions to states as in the national estimate so that the total emissions are the same in both inventories. These estimates are developed without reference to greenhouse gas inventories prepared by state governments, which may result in discrepancies in sectoral or total values due to different in methods or accounting. In addition to the GHGI state estimates, the EPA provides references to the independent inventories of 24 states and Washington, D.C. Of these, we find that eight produce a methane emissions inventory separate from a total greenhouse gases inventory based on CO­2-equivalents.

We partition our gridded posterior emissions estimates, excluding offshore emissions, to each of the 48 states in CONUS as described in section 2.8 and compare the results to the state GHGI and to inventories prepared by state governments. Figure 7 shows the results for the 29 states responsible for 90% of posterior CONUS anthropogenic emissions, and Table S1 shows the full results.

We find that the top 10 methane-producing states in the posterior estimate have emissions on average 38% larger than the GHGI and are responsible for 55% CONUS methane emissions. Oil and gas emissions have a disproportionate influence in these states, generating 47% of mean posterior emissions and 45% of the observed increase relative to the GHGI. In Texas, New Mexico, Louisiana, and Oklahoma, the oil and gas sector explains more than 60% of each state’s posterior emissions, with emissions increases concentrated in the Permian Basin (Texas and New Mexico), the Haynesville Shale (Louisiana), and the Anadarko Shale (Oklahoma). Livestock and landfills also play a significant role. Emissions in California and Iowa are dominated by the livestock sector, with much of the increase relative to the GHGI attributed to manure management (Section 3.1). Landfills account for 41% of posterior emissions in Illinois and 62% in Florida. Indeed, Florida is home to three of the ten largest landfills as reported by the GHGRP in 2019 (EPA GHGRP, 2022). Consistent with our sectoral analysis, the largest absolute negative posterior - state GHGI differences are found in coal-producing statesincluding Wyoming, West Virginia, and Pennsylvania.

We consider in more detail Texas and California, which are responsible for 21% and 6% of posterior CONUS anthropogenic emissions, respectively. Our posterior estimate for Texas is 6.3 (6.1 - 6.5) Tg a-1, a 69% increase from the state GHGI estimate of 3.7 Tg a-1. This increase is attributed almost entirely to the oil and gas sector, which accounts for 69% of posterior emissions compared to 56% in the state GHGI. The Permian basin alone explains almost 40% of Texas’ posterior emissions.

In California, we find posterior emissions of 2.1 (2.0 – 2.1) Tg a-1, 53% of which are in the San Joaquin Valley air basin. Our posterior emissions are a 33% increase from the state GHGI estimate of 1.5 Tg a-1, and a 32% increase from the independent estimate prepared by the California Air Resource Board (CARB) of 1.6 Tg a-1. Our posterior estimate is smaller than but consistent with the estimate of 2.4 ± 0.5 Tg a-1 found by an inversion of in situ observations in California from June 2013 to May 2014 (Jeong et al., 2016). We find good agreement with the sectoral partitioning in the state GHGI, the CARB inventory, and Jeong et al. (2016). Livestock explain 54% of emissions in our posterior estimate, 50% in the state GHGI, 54% in the CARB inventory, and 54% in Jeong et al. (2016), while landfills explain 25%, 23%, 21%, and 19% of emissions, respectively. We find slightly smaller relative contributions from oil and gas, which is responsible for 11% of emissions in our posterior estimate compared to 20%, 17%, and 18% in the state GHGI, the CARB inventory, and Jeong et al. (2016), respectively. This partitioning differs from that found in an inversion of observations from the 2010 CalNex aircraft campaign, where 30% of emissions were attributed to livestock, 38% to landfills, and 22% to oil and gas (Wecht et al., 2014).

We also compare our posterior emissions to independent state greenhouse gas inventories from Pennsylvania, Louisiana, Iowa, and Colorado, which are referenced by EPA (2022) and where the observing system provides a strong constraint (state averaging kernel sensitivity greater than 0.5). Our posterior agrees with the Pennsylvania estimate, but we find a source shift from fossil fuels (from 76% in the inventory to 63% in our work) to landfills (from 3% in the inventory to 16% in our work). We find that Louisiana’s state inventory is too low due to underestimated oil and gas emissions, while Iowa’s is too low due to underestimated livestock emissions, particularly from manure management (Section 3.1). Colorado’s state inventory is 65% larger than our work due to oil and gas emissions that are more than twice as large.

**3.4 Urban area emissions**

Urban areas are home to 81% of the U.S. population (U.S. Census, 2010) and are major sources of greenhouse gas emissions, including methane (Gurney et al., 2015; Hopkins et al., 2016). As urban populations grow (Seto et al., 2012), these emissions are likely to increase (Hopkins et al., 2016). Cities are well-positioned to address methane emissions through waste-reduction initiatives, leak-detection programs, and strategic contracts with landfill operators and gas utilities. Regulation by air pollution control districts can also aid urban emissions reduction efforts (Hopkins et al., 2016). C40, a performance-based coalition of over 100 mayors dedicated to climate change mitigation, recommends that cities target a 50% reduction in methane emissions by 2030 (C40, 2022). Numerous cities including New York City, Philadelphia, Washington, D.C., and San Francisco, are working toward these reductions through zero-waste programs. The U.S. Methane Emissions Reduction Action Plan intends to work with local governments to set up methane monitoring systems to identify and publicize information about municipal gas distribution leaks. The plan also challenges members of the U.S. Climate Mayors to prioritize pipeline abandonment or replacement (The White House, 2022).

We calculate posterior emissions for 95 urban areas across CONUS with 2010 populations over one million and averaging kernel sensitivities from our TROPOMI inversion greater than 0.2, providing the first comprehensive national analysis of urban methane emissions. Quantification of urban emissions depends significantly on the definition of urban area due to the presence of large emitters such as landfills on the periphery (e.g., Balashov et al., 2020; Plant et al., 2022). We follow Plant et al. (2022) and others in using the census designation to standardize the definition of urban areas across CONUS, and define urban areas using the U.S. Census Topographically Integrated Geographic Encoding and Referencing system (TIGER)/Line Urban Areas. These urban areas are responsible for almost a quarter of GHGI emissions when allocated using the gridded version of the EPA GHGI inventory ( Maasakkers et al.,2016). That gridded version does not include post-meter gas emissions, introduced in later versions of the GHGI, and which we distribute here d by population. Urban areas may also include emissions not related to population including oil/gas production and transport, and livestock, which we find account on average for 45% of urban area emissions. The remaining population-related emissions include landfills (40%), gas distribution (5%), post-meter emissions (5%), and wastewater (6%). , and other non-specific urban sources such as livestock and upstream oil and gas (45%).

Anthropogenic posterior emissions in these 95 urban areas total 6.0 (5.4 - 6.7) Tg a-1, 43 (29 - 60) % larger than the GHGI value of 4.2 Tg a-1. Individual urban area emissions, listed in Table S2, increase by an average of 47 (34 - 61) %. These increases are much larger than the 16 (12 - 19) % increase we find in total CONUS anthropogenic emissions relative to the GHGI. We are unable to attribute the emission increases to particular sectors due to source co-location within urban areas at the 0.25° × 0.3125° resolution of our inversion. However, given that landfills account for 40% of GHGI emissions in an average urban area and increase 52% relative to the GHGI, it is likely that they are responsible for a large fraction of the observed discrepancy. It is also likely that gas emissions, which represent less than 20% of GHGI emissions in an average urban area but explain between 32% and 100% of urban methane emissions (Plant et al., 2019; Floerchinger et al., 2021; Sargent et al., 2021), are significantly underestimated. The EPA added post-meter gas emissions of 457 Gg a-1 in their 2022 methane inventory, but this is insufficient to account for the observed discrepancy. Finally, recent studies have shown large underestimates of methane emissions from wastewater treatment (Moore et al., 2023; Song et al., 2023). City-specific variability prevents further attribution of urban emissions. Indeed, we find no correlation between the posterior increase in emissions and urban area population, population change from 2000 to 2010, population density, or surface area.

Figure 8 shows results for the top 10 methane-producing urban areas as ranked by posterior emissions from landfills, gas distribution, and wastewater. These 10 areas explain 35 (34 - 36) % of total urban posterior emissions. We find a mean increase relative to GHGI emissions of 74 (51 - 103) % that is not explained by population-dependent factors such as post-meter emissions or landfilled mass. We also compare our posterior emissions to municipal inventories from New York City and Philadelphia, the only available bottom-up urban methane emission estimates. Our emissions are more than twice as large but this likely results from our consideration of broader urban areas. Figure 8 further compares our results to 12 previous top-down studies published since 2015. Most focused on New York City or Los Angeles. Almost all the studies used larger definitions of urban area extent, with only Pitt et al. (2022) and Plant et al. (2022) using the U.S. Census designation. Most used aircraft or tower observations to infer emissions by inverting a CTM (Cui et al., 2015; Jeong et al., 2016; Cusworth et al., 2020; Pitt et al., 2022; Yadav et al., 2019, 2023). Kuwayama et al. (2019) used a mass balance approach, while others used observed methane to CO2 or CO ratios together with bottom-up inventories of these gases (Wong et al., 2015; Wunch et al., 2016; Plant et al., 2019). Plant et al. (2022) used the same approach with TROPOMI methane to CO emissions.

We find in general lower emissions than these previous studies, but this likely results from our restrictive definition of urban area. The only study that used aircraft data to estimate emissions within a U.S. Census Urban Area found 314 ± 96 Gg a-1 in New York City (Pitt et al., 2022), which is very similar to our estimate of 309 (241 - 417) Gg a-1. Plant et al. (2022) also used U.S. Census Urban Areas but relied on TROPOMI methane to CO ratios. They found slightly larger emissions in Atlanta and Philadelphia and much larger emissions in New York City, but their error bars spanned ranges that were almost twice as large as the derived emissions, limiting the utility of the comparison. Plant et al. (2019) also found larger emissions in New York and Philadelphia but used larger definitions of urban areas and produced similarly wide error ranges.

Our posterior Los Angeles emissions are much lower than previous studies shown in Figure 8. All these studies quantified emissions in CARB’s South Coast air basin, and we find a smaller but still significant low bias when we use an equivalent extent. We attribute much of the remaining discrepancy to a decrease of emissions with time. The 2013 closure of the Puente Hills Landfill, one of the largest landfills in CONUS, decreased methane emissions significantly (CARB, 2018; Yadav et al., 2019), which is not fully reflected in the estimates of Cui et al. (2015), Wong et al. (2015), or Wunch et al. (2016). Yadav et al. (2023) found that Los Angeles emissions decreased an additional 7% from January 2015 to May 2020. Their posterior estimate of 251 ± 5 Gg a-1 for 2019 is still larger than our value of 179 (171 – 193) Gg a-1.

**4 Conclusions**

We used TROPOMI atmospheric methane column observations for 2019 to optimize methane emissions at 0.25° × 0.3125° resolution over North America with focus on the contiguous US (CONUS). The high resolution of our inversion allows us to quantify emissions from individual landfills, states, and urban areas. We compared our results to the EPA Greenhouse Gases Inventory (GHGI) used for climate policy, including new state-level inventories, to Greenhouse Gases Reporting Program (GHGRP) inventories for individual landfills, and to other emission estimates.

Our optimization of methane emissions uses an analytical inversion of TROPOMI data with the GEOS-Chem chemical transport model at 0.25o×0.3125o resolution. The inverse solution (posterior estimate of emissions) is obtained through a reduced-rank approximation of the analytical minimum of a Bayesian cost function regularized by a prior emissions estimate from the EPA GHGI. The analytical solution characterizes the error and information content of the posterior emissions and supports the generation of an eight-member inversion ensemble. The Jacobian matrix needed for analytical solution at such high resolution on the continental scale is constructed by iterative approximation along the emissions patterns best informed by the prior emissions estimate and the observations. This decreases the computational cost of our inversion by an order of magnitude compared to conventional analytical approaches while optimally preserving its information content.

We find posterior anthropogenic methane emissions of 30.9 (30.0 - 31.8) Tg a-1 in CONUS, a 16 (12 - 19)% increase from the GHGI estimate of 26.7 Tg a-1, where ranges are provided by the inversion ensemble. Emissions for all sectors except coal increase relative to the GHGI. Most of the total increase is attributed to landfill emissions, which increase by 52%. Comparing our optimized emissions to those reported to EPA’s GHGRP by 73 individual landfills across CONUS, we attribute the observed discrepancy to EPA methodologies for estimating landfill emissions including overestimate of the decay rate of these emissions, too-high recovery efficiencies at facilities that collect landfill gas, and inadequate accounting of anomalous operating events such as gas leaks.

We took advantage of the high resolution of our inversion to quantify emissions for each of the 48 states in CONUS and compare to the newly available GHGI state estimates. We find a 38% average increase in the top 10 methane-producing states. Much of the discrepancy in these 10 states is due to increased oil and gas emissions, though livestock and landfills also play significant roles. Texas and California, the two largest methane-producing states, respectively emit 21% and 6% of total CONUS emissions in our posterior estimate. Emissions in Texas increase by 69% relative to the GHGI, almost entirely due to the oil and gas sector, including operations in the Permian basin, which explain almost 40% of all optimized emissions in the state. In California, we find a 33% increase from the GHGI and a 32% increase from an independent inventory prepared by CARB. Our sectoral contributions to methane emissions in California are consistent with those inventories including 54% from livestock, 25% from landfills, and 11% from oil and gas.

We also provide a first national analysis of urban methane emissions by calculating emissions for 95 urban areas across CONUS. We find total emissions of 6.0 (5.4 – 6.7) Tg a-1 across these urban areas, 43 (29 – 60) % larger than the spatially allocated GHGI value of 4.2 Tg a-1. 45% of urban emissions on average are from landfills and 10% are from gas distribution and post-meter emissions, but there are also large and variable contributions from oil/gas and livestock operations that are not specific to the urban population. The highest-emitting urban areas are New York City and Dallas. Our estimates of urban area emissions are generally lower than previous top-down studies but there is ambiguity as to the definition of urban area extent. Emissions in Los Angeles appear to have decreased significantly over the past decade.

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