Figure xx (solid lines) shows the prior GEOS-Chem – TROPOMI difference with respect to albedo, season, and latitude for the filtered data. We find no bias with respect to albedo or season and an aseasonal latitudinal bias. This bias has been noted and corrected previously by Turner et al. (xxxx), Maasakkers et al. (xxxx), and Zhang et al. (xxxx). We define a latitudinal correction term (ppb) for the GEOS-Chem – TROPOMI difference using the first-order polynomial

where is the degrees latitude. We find good agreement between the resulting prior GEOS-Chem output and the observations (R = 0.77).

We also conduct a suite of sensitivity tests to provide additional constraints on the error of the optimized emissions, which are summarized in section 2.6.

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

Houweling et al. (2017):

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

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

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

🡪 ask Daniel about this

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

🡪 ask Daniel about this

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

We optimize 3653 grid cells and obtain 558 DOFS in CONUS, with smaller total corrected areas in Mexico (462 grid cells and 88 DOFS) and Canada (380 grid cells and 56 DOFS).

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

The posterior solution depends on the choice of inversion parameters, including the prior emissions estimate, prior errors, the selection of observations, and the observing system errors. We solve the inversion for a range of

. The reduced-rank Jacobian matrix (section 2.6) optimizes emissions only in those grid cells that are informed by the observing system and defaults to the prior elsewhere.

together with the regularization factor (section 2.5) so that the prior term of the cost function evaluated at the posterior solution averages to 1 across all optimized grid cells (

We find large decreases in the three largest coal producing basins in CONUS: the Appalachian Basins, which stretch from Eastern Kentucky to Western Pennsylvania (); the Powder River Basin in Wyoming; and the Illinois Basin in Southern Illinois, Southern Indiana, and Western Kentucky.

Pennsylvania is the only state in the top 10 with a dominant contribution from coal emissions, which remain constant from the prior to the posterior. However, the co-location of coal and oil and natural gas facilities across Appalachia results in a large covariance between these source sectors in Pennsylvania, hindering accurate source attribution.

Lu et al. (2022) found slightly higher anthropogenic methane emissions of 36.9 (32.5 - 37.8) Tg a-1 over the same domain using a joint inversion of GOSAT and in situ observations for 2010 - 2017. However, their analysis of emissions trends showed posterior anthropogenic methane emissions for 2017 of 28.9 (28.4 - 29.5) Tg a-1, which are lower than our estimate.

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

EPA GHGI 9.4 total

EPA GHGI enteric fermentation 176.1 – 7.0

EPA GHGI manure management 58.7 – 2.3

Oil and natural gas emissions increase from 9.42 Tg a-1 to 10.49 (10.15 - 10.85) Tg a-1 across grid cells that explain 88% (80% - 92%) of prior emissions.

EPA GHGI 9.1 total

EPA GHGI natural gas 172.1 (includes 11.4 post-meter)

EPA GHGI petroleum systems 40.4

EPA GHGI stationary combustion 8.8

EPA GHGI abandoned oil and gas 7.0

Coal emissions decrease from 2.89 Tg a-1 to 1.46 (1.14 - 1.90) Tg a-1, with 96% (94% - 98%) of prior coal emissions optimized. The 49% (34% - 61%) decrease in emissions is consistent the 30% decrease in CONUS coal production since 2012 (USGS).

EPA GHGI 2.1 total

EPA GHGI coal mining 47.4

EPA GHGI abandoned underground coal 5.9

Landfill emissions increase by 27% (13% - 45%) from 5.65 Tg a-1 to 7.16 (6.40 - 8.21) Tg a-1 with 81% (62% - 90%) of prior emissions optimized.

EPA GHGI landfills 113.6 – 4.5

Current report includes post-meter

GOSAT produced an average of 25734 observations over North America per year from 2010 to 2017, providing an observational dataset with similar accuracy and precision but two orders of magnitude fewer measurements

Us 10.5 + 1.5 = 12

Lu 14.5 + 2.8 = 17.3

Deng 9.8 or 12.6

Open fire emissions are from the Global Fire Emissions Database version 4 (GFED4, van der Werf et al., 2017), termite emissions from Fung et al. (1991), and geological seepage from Etiope et al. (2019) scaled to the 2 Tg a-1 global emission magnitude given by Hmiel et al. (2020).

OVERESTIMATE

Colorado 1.338 vs. our 0.809 (0.740 – 0.861) and vs our prior of 0.930 . DOFS 0.59.

61% of the decrease is due to coal, which went from 0.176 to 0.102

Coal emissions decreased in the inventory from 0.326 to 0.073 from 2010 to 2019

Bulk of emissions are from oil and natural gas systems (0.801 or 60%), which is twice as large as our posterior estimate of 0.353, which is 44% of our total posterior emissions.

They have 0.296 (22%) from livestock while we have 0.232

0.073+0.801=874

0.102+0.353=455

New York 1.584 vs. our 0.431 (0.352 – 0.536) and our prior of 0.448 (DOFS 0.30)

Maybe no real constraint

No useful information on sectoral contributions in the inventory

UNDERESTIMATE

California 1.556. vs our 2.051 (1.944 – 2.124) and vs our prior of 1.764 DOFS 0.86

Our increase of 0.287 from livestock (78%).

California has no sectoral information in their inventory

Iowa 0.7724 vs. our 0.991 (0.943 – 1.017) and vs. our prior of 0.743 DOFS 0.75

Increase of 0.249 is 70% to livestock and 18% to landfills.

Iowa has no sectoral information in their inventory

Louisiana 0.375 vs our 1.188 (1.010 – 1.415) and vs. our prior of 0.721 DOFS 0.55

Increase of 0.334 from ONG, 72% of the observed increase (Remainder predominantly from landfills and other)

Louisiana has no sectoral information in their inventory

AGREE

Pennsylvania 1.278 vs our 1.2 (1.042 – 1.388) and vs our prior of 1.197 (DOFS 0.57)

Inventory attributes it to coal (37%) and ONG (39%) total 76%

We find coal 44% ONG 19% (total 63%), livestock 17% and landfills 17%

Originally 39% coal 34% ONG total 73% -- 11% landfills 14% livestock

AGREE and INCREASE Wisconsin 0.588 vs. our 0.575 (0.518 – 0.629) and vs. our prior of 0.519. DOFS 0.47

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| City | CH4:CO2 | CH4:CO | CO:CO2 | C2H6:CH4 | N |
| DC | 8.17  (6.20,10.95) | 1.13  (0.86, 1.60) | 8.58  (6.92, 12.28) | 0.033  (0.018, 0.057) | 7 |
| BLT | 6.07  (4.67, 6.94) | 1.27  (1.06, 1.56) | 4.72  (3.84, 5.86) | 0.029  (0.021, 0.037) | 5 |
| PHL | 5.17  (3.65, 6.10) | 1.02  (0.88, 1.30) | 5.16  (3.98, 6.29) | 0.026  (0.020, 0.030) | 5 |
| NYC | 7.21  (6.18, 8.56) | 1.16  (0.98, 1.58) | 6.84  (5.99, 7.75) | 0.017  (0.015, 0.020) | 14 |
| BOS | 4.24  (3.64, 4.70) | 0.84  (0.60, 1.01) | 4.74  (4.11, 5.33) | 0.022  (0.014, 0.036) | 8 |
| PVD | 4.16  (3.55, 5.21) | 0.48  (0.37,0.57) | 8.93  (5.75, 11.43) | 0.037  (0.015, 0.071) | 6 |

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | Total Emissions within "Urbanized Area" boundary | | | | | |
| Inventory | **DC** | **BLT** | **PHL** | **NYC** | **PVD** | **BOS** |
| CH4 (kg CH4/s) |  |  |  |  |  |  |
| Gridded EPA (April 2012) | 1.2 | 0.9 | 2.5 | 5.1 | 0.8 | 1.2 |
| EDGAR v4.2FT2010 (Annual 2010) | 3.4 | 2.4 | 7.1 | 15.3 | 1.0 | 3.2 |
| EDGAR v4.3.2 (April 2010) | 1.5 | 1.5 | 3.6 | 7.1 | 0.6 | 1.5 |
| CO2 (kg CO2/s) |  |  |  |  |  |  |
| Edgar v4.2FT2010 (Annual 2010) \* | 1173.9 | 1042.1 | 2571.5 | 5792.4 | 735.4 | 1481.3 |
| Edgar v4.3.2 (April 2010) \* | 1361.3 | 721.9 | 2515.1 | 6165.1 | 497.1 | 1603.0 |
| ODIAC2017 (April 2016) \* | 1055.0 | 1022.2 | 2062.3 | 3848.1 | 728.4 | 1360.1 |
| FFDAS2014b (April 2014) | 1782.1 | 1493.5 | 2746.5 | 5105.6 | 887.0 | 1802.2 |
| ACES (April 2014) | 1233.1 | 1041.4 | 2198.0 | 5195.1 | 738.8 | 1594.5 |
| CO (kg CO/s) |  |  |  |  |  |  |
| EDGAR v4.2.3 (April 2010) | 6.8 | 3.9 | 9.4 | 21.4 | 2.6 | 7.6 |
| NEI 2011 (April 2011) | 13.4 | 8.0 | 17.0 | 39.6 | 3.9 | 12.5 |

Sargent et al.

45 km radius of Boston

We present a top-down study of NG methane emissions from the Boston urban region spanning 8 y (2012 to 2020) to assess total emissions, their seasonality, and trends. We used methane and ethane observations from five sites in and around Boston, combined with a high-resolution transport model, to calculate methane emissions of 198 ± 47 Gg/yr, with 127 ± 24 Gg/yr attributed to NG losses. We found no significant trend in the NG loss rate over 8 y, despite efforts from the city and state to increase the rate of repairing NG pipeline leaks. We estimate that 2.5 ± 0.5% of the gas entering the urban region is lost, approximately three times higher than bottom-up estimates. We saw a strong correlation between top-down NG emissions and NG consumed on a seasonal basis. This suggests that consumption-driven losses, such as in transmission or end-use, may be a large component of emissions that is missing from inventories, and require future policy action.

City EDGAR v5.0 urban Emissions (kg CH4/s) EPA urban Emissions (kg CH4/s) Emission estimate – this work (kg CH4/s) 95% Confidence Interval

Atlanta 2.66 2.6 7.0 2.7, 15.2

Boston 1.4 1.4 5.1 1.9, 11.1

Washington 1.4 1.2 4.4 1.6, 9.8

Philadelphia 2.8 2.8 5.7 2.2, 12.2

New York 5.0 5.2 18.2 7.0, 39.2

Plant et al. (2022) combined TROPOMI methane : carbon monoxide ratios with carbon monoxide inventories to quantify methane emissions in eight geographically diverse cities in CONUS. We compare our results in the seven cities where TROPOMI constrains our posterior emissions and find that on average the Plant et al. (2022) estimate is 119% larger than ours. When we exclude Boston, where we have an urban averaging kernel sensitivity less than 0.1, the average relative difference is 103%. The magnitude of this difference is consistent with the difference compared to the carbon monoxide result in Plant et al. (2019), suggesting that the difference is attributable in part to errors in carbon monoxide inventories.

Lamb et al. (2016) and Jones et al. (2021) quantified methane emissions in Indianapolis using aircraft and in situ observations in inventory, mass balance, and inverse approaches. Lamb et al. (2016) found smaller emissions than ours in their observation-informed inventory and mass balance approaches, but larger emissions in their Bayesian inversion. Jones et al. (2021) found smaller emissions by almost half that were most consistent with the Lamb et al. (2016) mass balance approach. In all cases, the differences are insignificant.

Plant et al. (2019) inferred methane emissions from aircraft observations of ratios of methane to carbon dioxide and to carbon monoxide ratios over six East Coast cities. We compare our result to their carbon dioxide result, which relies on more accurate prior inventories, and find that their estimate is on average 75% larger than ours, with an insignificant difference only in New York City.

Pitt et al. (2022) conducted a Bayesian inversion of nine research flights over New York City and found of 313 (217 - 409) Gg a-1, where the parenthetical values represent the mean 1σ variability. This result is consistent with our estimate of 239 (165 - 341) Gg a-1.

Huang et al. (2016), Lopez-Coto et al. (2020), and Ren et al. (2018) quantified methane emissions over the Baltimore-D.C. metropolitan region using in situ and aircraft observations and found mean methane emissions between 212 and 468 Gg a-1. We find much smaller methane emissions of 29.9 (13.5 - 43.9) Gg a-1. The difference may be due to errors associated with coastal TROPOMI observations.

Cui et al. (2015), Jeong et al. (2016), Wunch et al. (2016), Yadav et al. (2019), and Cusworth et al. (2022) calculated methane emissions over Los Angeles for various periods beginning in 2007 and found mean methane emissions of between 333 and 413 Gg a-1. We find much smaller emissions of 90.5 (81.9 -106.8) Gg a-1, a decrease relative to the prior of 133 Gg a-1.

Fairley and Fischer (2015), Jeong et al. (2016, 2017), and Guha et al. (2022) quantified methane emissions over San Francisco and found mean emissions between 222 and 245 Gg a-1. We find much smaller emissions of 17.1 (12.6 – 24.1) Gg a-1.

Sargent et al. () found methane emissions of 198 (151 - 245) Gg a-1.

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

EPA GHGI 9.4 total

EPA GHGI enteric fermentation 7.0

EPA GHGI manure management 2.3

“A recent global study suggests a similar underestimation for manure management in a bottom-up inventory. Based on published data on field-scale measurements of GHG emissions, Owen and Silver [2015] report that predicted CH4 emissions by the Intergovernmental Panel on Climate Change Tier 2 method are lower than the mean estimates using the field measurements for most manure management practices”

https://onlinelibrary.wiley.com/doi/abs/10.1111/gcb.12687

[define observing system]

Inverse studies summary paragraph

Papers to cite

NA inversions

* Miller et al. 2013 – Lagrangian inversion of observations from towers and aircraft, analyzes footprints for each of 12,694 observations, geostatistical inversion (no prior), 1deg 1deg
* Wecht et al. 2014 – SCIAMACHY, adjoint
* Turner et al. 2015 – GOSAT, GMM, EDGAR
* Janardanan et al. 2017? – don’t actually optimize emissions, just look at the differrence
* Bruhwiler et al. 2017?
* Sheng et al. 2018?
* Lan et al. 2019?
* Maasakkers et al. 2021 – GOSAT, GMM, EPA GHGI

High resolution regional inversions

* Wecht et al. 2014 – analytical inversion over western North America and Pacific? (157 grid cells at 0.5 degrees) (Spatially resolving methane emissions in California, ACP)