

We thank both reviewers for their comments.

Reviewer 1

General comments

This is an excellent, well-crafted and important paper. The findings have significant implications for our understanding of US, Canadian and Mexican methane emissions. They also draw attention to knowledge gaps – for example in quantification of methane fluxes from wetlands. The methodology is detailed and convincingly performed: assuming there are no unexpected errors of computation the results should be sound and uncertainties appropriate, given the limitations of the data. The work should be published after minor revisions.

Specific comments

P2 Line 5 – Very dated flavour to this - maybe add some more up to date work later than Kirschke et al. 2013 - for example Saunio et al papers, and also perhaps recent measurements of global growth.

We have added Saunio et al. (2020) as a reference and described the global growth rate.

Global methane concentrations have increased by a factor 2.5 compared to preindustrial times Hartmann et al. (2013) and by 7.1 ppb⁻¹ since 2007 (with the rate peaking above 10 ppb a⁻¹ in 2014 and 2015) after a period of stability in the early 2000s (https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/, last access: 20 December 2020). Major emission source sectors include wetlands (the main natural source), livestock, the fossil fuel industry, and waste management (Kirschke et al., 2013; Saunio et al., 2020).

P2 L20 – P3 L7. These paragraphs have a sense of being written a long time ago and only given a quick brush-over update with the addition of brief cursory mentions of the work by Lan et al. 2019 and Bruhwiler et al. 2017. For a non-North American readers (this is a European journal), it would help to give more discussion in these paragraphs to these two recent and important studies.

We added more context on the difference between Turner et al. (2016) and Bruhwiler et al. (2017) and added more information on the Lan et al. (2019) study:

By contrast, Bruhwiler et al. (2017) found from an ensemble of inversions using surface and satellite observations that North American emissions had been flat for 2000-2012 and that without an inverse model short-term trends can appear to be present in the GOSAT data because of interannual transport variability, choice of background, and the seasonal sampling bias of GOSAT.

Using 2006-2015 surface and aircraft observations over the US and Canada, Lan et al. (2019) found a 0.7 ± 0.3 % a⁻¹ increase in total US emissions and a 3.4 ± 1.4 % a⁻¹ increase in oil/gas emissions based on stations in Oklahoma, North Dakota, and Texas. They also reported an increase in the ethane:methane emission ratio which could lead to an overestimate of the oil/gas methane trend as inferred from the ethane trend.

P3 L33 – P4 L1. How much does the cloud cover problem affect GOSAT sensing of wetland emissions? In the equatorial tropics the cloud cover is essentially 100% all through the rainy seasons so it's hard to argue that the cloud-free periods are representative: indeed, they may be warmer. In North America however, with weather system passages, it could be argued that cloud-free episodes are disproportionately in cool clear weather when moisture (and methane) advection is least. Thus GOSAT may disproportionately observe low flux periods.

Through the use of 6 years of data and the ability of the inversion to attribute downwind concentrations to emitting areas with cloud cover during observation, we feel confident the inversion can provide the presented constraints on wetlands. Because of the magnitude of the coastal wetland emissions in our prior, the averaging kernels for these areas are large as later discussed and shown in Figure 3.

P4 L11 Table 1 – please can this table also estimate the Soil Sink, which is passed over in this Table and not well discussed in the paper as a whole (see also P9 L15).

We have added the soil sink as a footnote to the table to be able to add that it is not optimized:

The soil sink is 3.6 Tg a^{-1} for the inversion domain (Fung et al., 1991) and is not optimized in the inversion.

P4 L15-18 – For the priors, the Canadian data are from a business consultancy ICF, and are not from a governmental or peer reviewed source. Thus the use of this data set needs to be defended. Why is this ICF inventory preferred to the Canadian governmental data – does this choice of prior imply that something is assumed to be wrong with the Canadian UNFCCC submission?

We added an explanation for this:

The ICF inventory was used as the basis for the Sheng et al. (2017) gridded emissions as it provides a detailed breakdown of sources using methodology similar to the EPA GHGI.

P4 L5 – Is Fung et al. 1991 the most recent information about termites?

The Fung et al. (1991) termite emissions are in the current standard simulation methane in GEOS-Chem, their global total of 12 Tg a^{-1} fits within the range reported by Saunio et al. (2020) of 9 (3-15) Tg a^{-1} . We added the global total to aid in the interpretation.

... termite emissions are from Fung et al. (1991) with a global total of 12 Tg a^{-1} .

P5 L1 – the authors will be aware of the recent publications challenging these estimates of seepage and implying they are much too high. It would be useful to comment on why the higher values are chosen, as they are fairly hard to defend given the ice core evidence.

The global total of the seepage emissions we use is 5 Tg a^{-1} , under the 5.4 Tg a^{-1} maximum proposed for the pre-industrial age by Hmiel et al. (2020). We added the global total and added a comparison to Hmiel et al. (2020) to clarify this.

We use geological seepage emissions compiled from literature on both point sources (Etiope, 2015; Kvenvolden and Rogers, 2005) and areal seepage (Kvenvolden and Rogers, 2005; Etiope and Klusman, 2010) as described in Maasakkers et al. (2019) with a global total of 5 Tg a^{-1} , under the 5.4 Tg a^{-1} maximum proposed for pre-industrial times by Hmiel et al. (2020) based on ice core measurements.

P5 L18 – for those of us not familiar with this correction, maybe add a little justification of the term – does this reflect the Brewer-Dobson circulation? How much impact on North America is there from 1. the polar vortex downwelling of low methane air, and 2. major convective systems upwelling in the Gulf of Mexico region?

We added an explanation for the biases and the impact of the polar vortex.

*Following Maasakkers et al. (2019), we correct the GEOS-Chem simulation of GOSAT columns for a latitudinally and seasonally variable background bias likely caused by the extratropical stratosphere (Bader et al., 2016; Saad et al., 2016; Stanevich, 2018). **The bias is common in atmospheric models and caused by excessive meridional transport in the stratosphere (Patra et al., 2011) and in particular in the seasonal polar vortices (Zhang et al., 2020b).***

P6 Table1 Open fires – ‘natural’ – though most are human lit. I’m surprised Canada is only 60% of the CONUS: it has some pretty big fires! Seeps number seems too big. Gas distribution in Mexico 0 and production in Mexico 01.? The ICF and IMP data sources are rather old – I wonder if they ‘look back’ a longish way. Are these priors valid for 2010-15? (NOTE – these comments were written before seeing Fig. 4)

As mentioned with respect to comment P5L1 the global seeps emissions from the database we used are rather small. We have added a footnote to the table to mention all values are rounded and changed all zeros to ‘<0.1’ in the table.

All values in the table are rounded to one decimal.

The ICF and IMP comparisons to the UNFCCC data in Section 2.2 are with respect to the latest information available for that time period (UNFCCC, 2020). The years the inventories were based on (2010 and 2013 for Mexico and Canada respectively) are both within the simulation period.

P10 L8-9 q10 and CH₄:CO₂ discussion is interesting and might be worth extending a little as it’s important.

We added some additional discussion:

The large uncertainty range is driven by the inversion ensemble member without seasonal correction. Based on the root mean square error and spatial correlation, our inversion results are most consistent with the WetCHARTs ensemble members that use GlobCover wetland extent, a $q_{10} = 2$ value for the factor increase in the CH₄ to CO₂ emission ratio per 10 K temperature increase (a critical quantity for determining the sensitivity of wetland CH₄ production to temperature (Yvon-Durocher et al., 2014; Bloom et al., 2016)), and global scaling at the low end or middle of the range (global wetlands emission range 125-166 Tg a⁻¹). A value of $q_{10} = 2$ is approximately equivalent with the average CH₄ to CO₂ temperature sensitivity reported by Yvon-Durocher et al. (2014) based on meta-analyses, which indicates that anaerobic CH₄ respiration is substantially more sensitive to temperature relative to overall CO₂ respiration rates.

P11 L108 are significant and the Figure is important and very useful. P12 L8 – 30% lower. Has there also been a reduction in deliberate venting and inefficient flaring?

We have clarified that these changes are not progress over time but rather revisions to the emission factors/data used in the inventory’s calculation.

Our scaling factors to the EPA GHGI are for the 2012 emissions as reported by EPA (2016) and used in the inversion as prior estimates. More recently, EPA (2020) updated its methodology for estimating emissions and applied it to a reanalysis of emissions from previous years including 2012. Changes for 2012 emissions are important for some oil/gas subsectors, as shown in Figure 5. Gas production emissions in 2012 are lower by 19% in the updated GHGI because of a downward correction to emissions from gathering and boosting stations. Oil production emissions in 2012 are 30% lower in the updated EPA GHGI because of previous faulty double-counting of wells. Our correction factor from the inversion increases oil production 10 emissions by a factor 1.9 (1.7-2.3) and natural gas production emissions by a factor 1.5 (1.4-1.6) relative to the updated 2012 GHGI from EPA (2020). The updated GHGI emissions from natural gas processing in 2012 are 55% lower than previously reported, but our inversion finds them to be higher. Our correction factor from the inversion increases gas processing emissions by a factor of 2.9 (2.6-3.1) relative to the updated GHGI.

P13 Table 2 is very valuable. P14 L11 – Aha!- at last a brief further comment on Lan et al. It would help to mention Lan et al rather more in this paper. From the European perspective both the Lan and Bruhwiler studies seem to be important and well worth more discussion. We have extended the discussion of both papers in the introduction as discussed related to comment P2 L20 – P3 L7.

P14 L16-17. This is very depressing that landfill emissions are not decreasing. That surely implies a regulatory failure of some magnitude?

Not necessarily, the change may have been too small or wide-spread and mixed with other emissions to be picked up by the inversion, we rephrased the sentence:

*On the national scale, however, **the inversion does not detect decreasing emissions from landfills or coal mining.***

Additionally, landfill emissions had already decreased by 31% in 2010 compared to 1990 based on the GHGI.

P14 L18-31 Likewise this paragraph seems to imply that there has been little improvement in practice and technology over the period of study and that all emission changes are due to production shifts (for example to the Marcellus). Is this really so? Have leak and vent reduction efforts and technological improvements really had so little impact?

The increase shown here is much smaller than the relative increase in production over the same area suggesting that the leak rate did in fact decrease, but emissions did increase more than suggested by the EPA GHGI. We added a caveat:

*Most of our increase is driven by the Marcellus Shale area in the Northeast US, amounting to 130 (20-190) Gg α^{-1} α^{-1} . This area covering Pennsylvania, Ohio, and West Virginia has seen a large increase in natural gas production driven by unconventional drilling. Natural gas production in the area increased by a factor 7.8 between 2010 and 2015, contributing 22% of US natural gas production in 2015 (EIA, 2020a). **If the 130 (20-190) Gg α^{-1} α^{-1} increase is entirely due to gas production, it would only amount to 15 (2-21)% α^{-1} relative to the mean posterior gas production emissions in the area (0.89 Tg α^{-1}) indicating that the leakage rate did decrease over the time period.***

P14 last paragraph and onto p15 – Canada is trying hard to reduce methane emissions – is the emission decrease really only due to production shifts and not to the regulatory and technical changes? Has government action really had no impact?

Because of multiple changes in Canada it is difficult to attribute the apparent decrease that we see, we have added context on Canada in the manuscript:

In Canada, gas production has been stable while oil production from oil sands has increased, but the number of wells has decreased and efforts have been made to reduce emissions (Natural Resources Canada, 2020).

P15 L34 – Separating Urban emissions from wetlands is a global problem as so many cities are in deltas. This is probably beyond what is possible from satellites and needs in situ and aircraft field data - both isotopic sampling and vehicle based mapping.

We agree that additional data from either other platforms or newer satellites (TROPOMI) provide exciting prospects to aid the disentanglement of wetland and urban emissions.

CONCLUSION. This is an important paper, well written and with significant findings. It should be published after minor revisions

Reviewer 2

General comments

This paper presents a GEOS-Chem 2010-2015 inversion of CH₄ sources over North America. Sectoral emissions and their trends are optimized using a state defined by a so-called Gaussian Mixture Model (GMM), published earlier. Emissions are constrained by GOSAT observations. From an ensemble of inversions, it is found that emissions from the oil and gas sector are higher than in bottom-up reporting. Also, a slight positive trend of 0.4% per year in US anthropogenic methane emissions is derived. The paper is well written, referencing is adequate, and the results are compared to previous studies. In that respect, the paper is a valuable contribution and deserves publication. However, I also find that the paper cleverly hides some of the bottlenecks in the set-up. I mention two major issues that require further attention/discussion.

First, the inversion is driven by 156110 GOSAT observations in the 2010-2015 timeframe. On page 9, line 29, the authors write that the mean squared difference with GOSAT is reduced by only 3.5% by optimizing the emissions. This relatively small improvement is ascribed to the already good fit using prior emissions, due to the optimized boundary conditions (global Geos-Chem inversion in which emission are optimized by GOSAT observations). This implies that the GOSAT observations are used twice: (1) in the global observations to set the boundary conditions, and (2) in the regional inversion using these boundary conditions. Although I think this is not a major issue, some mention of this drawback is needed (other approaches have been developed to circumvent this issue, e.g. Rödenbeck, C., Gerbig, C., Trusilova, K., and Heimann, M.: A two-step scheme for high-resolution regional atmospheric trace gas inversions based on independent models, *Atmos. Chem. Phys.*, 9, 5331– 5342, <https://doi.org/10.5194/acp-9-5331-2009>, 2009.)

We have added an additional explanation of the used boundary conditions in section 2.3:

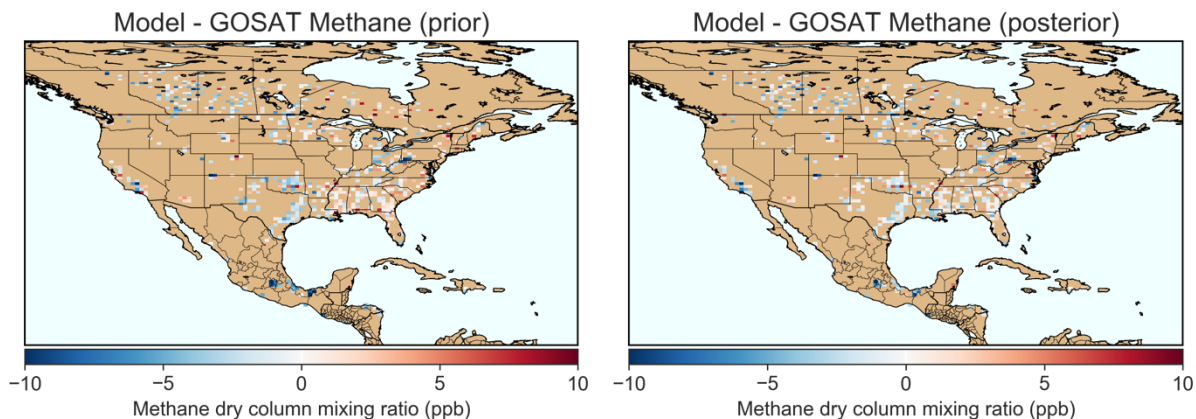
*Three-hourly boundary conditions at the edges of the nested domain are from the $4^{\circ} \times 5^{\circ}$ posterior model simulation of Maasakkers et al. (2019), which provides an unbiased fit to the global GOSAT data. **That posterior simulation includes some information from GOSAT data over the North America domain, which were used (along with the more abundant data outside that domain) in the global inversion; but the main consideration here is to avoid bias in boundary conditions that would otherwise affect the North American inversion.***

Next to this, I was surprised by the large increase in correlation with independent surface observations (r-squared increases from 0.58 to 0.81). Presumably, emissions (and their associated seasonal cycles) are adjusted such that temporal correlations increase. However, the authors provide remarkably little information. In fact, we do not get any information (other than the numbers above) about the ability of the posterior model to simulate GOSAT and surface observations. I also wonder why both data-sources are not assimilated together. Likely there is an unmentioned bias. To remedy this, I suggest that the authors present metrics/figures concerning prior/posterior mismatches with assimilated and unassimilated data.

We have added additional metrics in the manuscript:

The posterior emissions when implemented in GEOS-Chem reduce the mean squared difference with GOSAT observations by 3.5%. This overall reduction in error small because random errors in individual observations are large and because the background is already captured well in the prior simulation through the optimized boundary conditions. The main improvements are found over areas where the averaging kernels are large (Figure 3). For data with averaging kernel sensitivities greater than 0.1, the mean squared difference is reduced by 6.1% and the correlation increases from 0.62 to 0.64. We independently evaluated the posterior estimate by comparison to in situ methane concentrations from surface sites reported in the GLOBALVIEWplus CH₄ ObsPack v1.0 data product compiled by the NOAA Global Monitoring Laboratory (Cooperative Global Atmospheric Data Integration Project, 2019). Compared to the prior simulation (Reduced major axis (RMA) slope: 0.69, $r^2 = 0.39$), the posterior simulation (RMA slope: 0.69, $r^2 = 0.45$) does not degrade the comparison with these data and improves the correlation. The spatial coefficient of determination between the time-averaged GEOS-Chem and NOAA data increases from $r^2 = 0.58$ with the prior emissions to $r^2 = 0.81$ with the posterior emissions, representing an improvement in our ability to fit observed patterns.

The NOAA data are not assimilated here to be able to use them as an independent evaluation of the inversion results, combining GOSAT and NOAA is explored in Lu et al. (2020). Here, we have included a comparison between the model and GOSAT on the model resolution for cells with an averaging kernel larger than 0.1 and at least 5 observations below to provide additional context.



Second, the authors mention that they developed an analytical optimization, based on 1200 model simulations. This makes it easy to explore sensitivities once the simulations have been performed. In an analytic inversion framework, the calculation of the posterior co-variance matrix is possible. However, on page 11, the authors only present a metric that indicates how well the observations constrain the emissions of particular emissions (actually, I would move this part to the method section). One aspect is missing in this analysis: It would be interesting to know what is the co-variation of total wetland emissions and anthropogenic emissions, because natural emissions (are reduced from 15.7 to 11.8 Tg per year) can only to some extent be separated from anthropogenic emissions (which increase from 28.7 to 30.6 Tg per year). The co-variance matrix would inform on this co-variation (how well can you separate these emissions?), as well as on

the uncertainty reduction associated with individual emissions. Although the “sensitivity” inversions provide useful information, I think their range remains always somewhat subjective, depending on the choices made. For instance, is the range in gamma values (0.1, 0.5, 1.0) logical? Why is there no sensitivity for different/perturbed boundary conditions? In that sense, the posterior co-variance of a particular inversion is a useful additional metric that should be reported, when its calculation is feasible (which I guess is the case, given the fact that the averaging kernel matrix is explored).

The uncertainties derived from the posterior error covariance matrix for Bayesian inversions are often overly optimistic because they assume fully random errors on the many observations used whereas both model and satellite errors are correlated. This is worse when multiple state vector elements are summed. Even for the results that rely on one Gaussian such as Mexico City, the posterior error is rather small. Based on the posterior error covariance of the base inversion, the uncertainty on the scaling factor would be 1.56 ± 0.12 , a range much narrower than the 1.56 (1.31-2.20) reported based on the ensemble. We added the reasoning behind this in section 2:

The posterior error covariance matrix from the inversion underestimates the actual uncertainty in the results because of the assumption of fully random observational errors (Jacob et al., 2016). Therefore we use the range of results from the inversion ensemble as a better measure of uncertainty (Heald et al., 2004).

The use of the covariance matrix to study the correlation between source sectors is a very interesting suggestion that we implemented in the manuscript, we also added a definition for the mapped posterior error covariance matrix and made an improvement to the calculation of the averaging kernel sensitivities that leads to slightly different values in the table:

The summation matrix (\mathbf{W}) weighs the relative contribution $w_{i,k}$ of sector/subsector i to the total emission in Gaussian k in the prior inventory:

$$\begin{aligned}\mathbf{A}_{\text{sub}} &= \mathbf{W}\mathbf{W}^* \\ \hat{\mathbf{S}}_{\text{sub}} &= \mathbf{W}\hat{\mathbf{S}}\mathbf{W}^T\end{aligned}$$

Here $\mathbf{W}^ = \mathbf{W}^T(\mathbf{W}\mathbf{W}^T)^{-1}$ is the generalized pseudo-inverse of \mathbf{W} , and \mathbf{A}_{sub} (with diagonal elements $a_{i,i}$) and $\hat{\mathbf{S}}_{\text{sub}}$ are the averaging kernel matrix and posterior error covariance matrix mapped to the different subsectors. $a_{i,i} = 1$ means that the inversion can fully constrain the national total for that emission category, independent of the prior estimate, while $a_{i,i} = 0$ means that the inversion provides no information and the estimate cannot depart from the prior. The off-diagonal elements of $\hat{\mathbf{S}}_{\text{sub}}$ measure the error correlation in the posterior solution for different subsectors, and this is important to diagnose whether we can optimize different subsectors independently. The diagonal elements of $\hat{\mathbf{S}}_{\text{sub}}$ estimate the error variance in the posterior solution for individual subsectors but that estimate is too small because it assumes that the observations are independent and identically distributed (IID condition) (Brasseur and Jacob, 2017). We prefer to estimate the error in the posterior solution from the results of the inversion ensemble, as shown in Table 2.*

Our posterior estimate for the mean 2010-2015 CONUS anthropogenic source is 30.6 (29.4 - 31.3) Tg a^{-1} , where the best estimate is from the base inversion and the range is from the inversion ensemble. The 2012 emission total from the EPA GHGI 25 (EPA, 2016) used as prior estimate in our inversion is 28.7 Tg a^{-1} , with an uncertainty range 26.4 - 36.2 . We find limited posterior error correlation ($r = 0.33$) between the posterior anthropogenic and natural emission totals. Examining the contributions from different sectors, our best

posterior estimates for landfills and livestock are within 5% of the GHGI, and coal emissions are 6% higher. Oil/gas emissions total 11.1 Tg a^{-1} in our base inversion, 22 (12-32)% higher than the GHGI, and driven by oil/gas production as seen for example in Texas, Oklahoma, and offshore in the Gulf of Mexico. Our national estimates for the emissions from oil and gas production are 3.1 ($2.7\text{-}3.6$) and 5.4 ($4.9\text{-}5.9$) Tg a^{-1} , respectively, as compared to 2.3 and 4.4 Tg a^{-1} in the GHGI. **The posterior error covariance between wetland emissions and both oil production ($r = 0.02$) and gas production ($r = 0.04$) are low, showing that this increase is independent of the large decrease in wetland emissions.**

Finally, I include an annotated pdf which contains minor comments and suggestions. These comments are addressed below.

Specific comments (adopted from PDF comments)

P1L7 Strange acronym!

The acronym refers to ‘Greenhouse Gas Inventory’ and is commonly used in the literature, we clarified this in the main text.

P2L1 My first impression is that some words about the BCs are missing.
See response to ‘general comments’.

P2L10 What then is the "I". Inventory I guessed?
We clarified the acronym:

*U.S. Greenhouse Gas Emissions and Sinks (**Greenhouse Gas Inventory**, GHGI)*

P2L12 Since we use priors in inversions, this is not totally true.
We rephrased the sentence:

*Measurements of atmospheric methane, including from satellites, can be used through inverse modeling to provide an **evaluation** of these emission estimates (Streets et al., 2013; Jacob et al., 2016).*

P3L3 formatting seems random, e.g. "a" is italic below.
We corrected the italic formatting of ‘a’ on line 6.

P3L6 Would it be instructive to add a similar picture indicating the amounts of observations. I can imagine that observations get sparser towards 60N. Unclear is the "counter-direction" stripe pattern. Maybe an explanation is needed here?
We clarified the pattern in the observations and their repeat time in the caption of Figure 1:
The apparent north-south tracks that are GOSAT's default mode observations with a repeat cycle of 3 days, while the off-track data are target mode observations.

P9L31 This makes me wonder if you are not using the observations multiple times: (1) for the global inversion, setting up the BCs (2) for local constraints.
See response to ‘general comments’.

P10L2 Interesting result. Given the fact that the GOSAT improvement is small, this is quite remarkable. Apparently there are seasonal adjustments driven by GOSAT, that

improve the r^2 . But I am also interested in biases. Why are these observations not also assimilated? This sounds that this could be done without any problems. Given the fact that the GOSAT improvement is small, this is a remarkable result.

See response to 'general comments'.

P10L14 But if you use the same observational data, this detail merely comes from better resolution.

We clarified this:

*The large-scale correction patterns revealed by the inversion are similar to those of the coarse ($4^\circ \times 5^\circ$) global inversion reported by Maasakkers et al. (2019), which used the same prior estimates, but we have much more detail here **allowed by the higher resolution of the inversion.***

P10L19 rewrite: i read this sentence a couple of times, without success.

We rephrased the sentence:

A narrow uncertainty range does not necessarily reflect confidence in the inversion results. For small source sectors, it may also be due to insufficient information from the observations so that the optimization is unable to depart from the prior estimate. This can be determined using the averaging kernel sensitivities, as will be done below for the US (see Table 2).

P11L4 Here I lack information about the Gaussians used. Are there Gaussians centered around Mexico city?

We added this information:

*We also find 56 (31-120) % higher emissions over Mexico City, **which is optimized by a single Gaussian covering five grid cells. The difference is** attributed to wastewater based on the EDGAR spatial patterns. Compared to EDGAR v4.3.2 **a recent gridded inventory for Mexico (Scarpelli et al., 2020)** and the Mexico City Secretariat of Environment (SEDEMA, 2018) air quality emission inventory predict lower emissions from wastewater (68 versus 259 Gg a^{-1} **in the SEDEMA inventory**) but much higher landfill emissions (222 versus 1 Gg a^{-1}) indicating that our higher emission estimate may be related to landfill emissions being misallocated in EDGAR v4.3.2.*

P11L23 In theory, this could also be applied to co-variances between sectors. This would be very informative.

See response to 'general comments'.

P11L28 This range is quite subjective because it is determined by your (arbitrary) choice of sensitivity simulations.

See response to 'general comments'.

P13T2 Theoretically, the error reduction in the sum should always be smaller than the individual sub-sectors. I am not sure if this also holds for the a_i 's.

We added a clarification on the calculation of a_i for the sum of emissions:

We also calculate a_i for the sum of US anthropogenic emission categories and find emissions are 53% informed by the observations, with less information for individual sectors/subsectors.

P16L18 It would be interesting to know how wetland (natural) and oil/gas (anthropogenic) emissions are correlated in the posterior. Here it looks like lower wetland emissions go hand-in-hand in higher oil-gas emissions.

See response to ‘general comments’.

P25F3 But these are the Gaussians, right? So add: projected on the model grid.

We have changed the caption:

*Mean 2010-2015 posterior methane emissions **projected from the 600-member Gaussian mixture to the 0.5°x0.625° model grid** and comparison to the prior estimate. Results are from the base inversion. The bottom right panel shows the averaging kernel sensitivities **projected to the model grid** (diagonal elements of the averaging kernel matrix).*

We made some other minor textual improvements to improve the flow of the manuscript after incorporation of the feedback from the reviewers.

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2010-2015 North American methane emissions, sectoral contributions, and trends: a high-resolution inversion of GOSAT satellite observations of atmospheric methane

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Abstract. We use 2010-2015 GOSAT satellite observations of atmospheric methane columns over North America in a high-resolution inversion of methane emissions, including contributions from different sectors and ~~long-term trends~~ their trends over the period. The inversion involves analytical solution to the Bayesian optimization problem for a Gaussian mixture model (GMM) of the emission field with up to $0.5^\circ \times 0.625^\circ$ resolution in concentrated source regions. Analytical solution provides a closed-form characterization of the information content from the inversion and facilitates the construction of a large ensemble of solutions exploring the effect of different uncertainties and assumptions in the inverse analysis. Prior estimates for the inversion include a gridded version of the EPA Inventory of U.S. Greenhouse Gas Emissions and Sinks (GHGI) and the WetCHARTS model ensemble for wetlands. Our best estimate for mean 2010-2015 US anthropogenic emissions is 30.6 (range: 29.4-31.3) Tg a⁻¹, slightly higher than the gridded EPA inventory (28.7 (26.4-36.2) Tg a⁻¹). The main discrepancy is for the oil and gas production sectors where we find higher emissions than the GHGI by 35% and 22% respectively. The most recent version of the EPA GHGI revises downward its estimate of emissions from oil production and we find that these are a factor 2 lower than our estimate. Our best estimate of US wetland emissions is 10.2 (5.6-11.1) Tg a⁻¹, on the low end of the prior WetCHARTS inventory uncertainty range (14.2 (3.3-32.4) Tg a⁻¹) and calling for better understanding of these emissions. We find an increasing trend in US anthropogenic emissions over 2010-2015 of 0.4% a⁻¹, lower than previous GOSAT-based estimates but opposite to the decrease reported by the EPA GHGI. Most of this increase appears driven by unconventional oil/gas production in the eastern US. We also find that oil/gas production emissions in Mexico are higher than in the nationally

reported inventory, though there is evidence for a 2010-2015 decrease in emissions from offshore oil production.

1 Introduction

Methane is the second most important greenhouse gas in terms of radiative forcing (Stocker et al., 2013). [Global methane concentrations have increased by a factor 2.5 compared to preindustrial times Hartmann et al. \(2013\) and by 7.1 ppb \$\text{a}^{-1}\$ since 2007 \(with the rate peaking above 10 ppb \$\text{a}^{-1}\$ in 2014 and 2015\) after a period of stability in the early 2000s \(https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/, last access: 20 December 2020\).](#) Major emission source sectors include wetlands (the main natural source), livestock, the fossil fuel industry, and waste management ([Kirschke et al., 2013](#)) ([Kirschke et al., 2013](#); [Saunio et al., 2020](#)). Individual countries report their anthropogenic emissions to the United Nations Framework Convention on Climate Change (UNFCCC) using methods prescribed by The Intergovernmental Panel on Climate Change (IPCC) (United Nations, 1992; IPCC, 2006). The reports use “bottom-up” methods, where activity data (e.g., heads of cattle) are combined with emission factors (e.g., emission per head of cattle) to estimate total emissions. US emissions are calculated and reported in this manner by the Environmental Protection Agency (EPA) in its annual Inventory of U.S. Greenhouse Gas Emissions and Sinks ([Greenhouse Gas Inventory](#), GHGI) (EPA, 2020). Measurements of atmospheric methane, including from satellites, can be used through inverse modeling to provide an [independent-comparison-evaluation](#) of these emission estimates (Streets et al., 2013; Jacob et al., 2016). Here, we evaluate 2010-2015 North American emissions by inversion of data from the Greenhouse Gases Observing Satellite (GOSAT), which measures methane concentrations at high precision by solar backscatter in the shortwave infrared (SWIR) (Butz et al., 2011; Buchwitz et al., 2015; Kuze et al., 2016). We take the gridded version of the EPA GHGI (Maasakkers et al., 2016) as prior estimate for the inversion, enabling us to use the inversion results to evaluate the GHGI and guide improvements in its representation of emission processes.

Several inverse studies using observations of atmospheric methane have found higher US emissions than reported in bottom-up inventories. Miller et al. (2013) estimated methane emissions 50% higher than the EPA GHGI based on 2007-2008 surface and aircraft observations. They attributed this difference to fossil fuel extraction. Wecht et al. (2014) estimated 40% higher livestock emissions using 2004 data from the SCIAMACHY satellite instrument (Frankenberg et al., 2006). Turner et al. (2015) found anthropogenic emissions to be 50% higher than the EPA GHGI by inversion of 2009-2011 GOSAT data, attributing the difference to a combination of oil/gas and livestock emissions. Janardanan et al. (2017) found 28% higher anthropogenic emissions over North America based on 2009-2012 GOSAT data. All these studies used the global gridded EDGAR inventory (European Commission, 2011; Olivier and Janssens-Maenhout, 2012; Janssens-Maenhout et al., 2019) as prior estimate, but the EDGAR spatial distributions have large errors that affect inversion results and their interpretation (Maasakkers et al., 2016). Alvarez et al. (2018) used surface and aircraft data in oil/gas fields to find emissions from oil/gas production 60% higher than in the EPA GHGI.

There has also been substantial debate as to the contribution of North American emissions to the rising global methane trend since 2007. Hausmann et al. (2016) proposed an increase in US gas production as explanation for the 2007-2014 methane and ethane concentration trends at the Zugspitze mountain site in southern Germany. Turner et al. (2016) found a $2.5\% \text{ a}^{-1}$ increase in US emissions for 2002-2014 on the basis of GOSAT and surface methane data. By contrast, Bruhwiler et al. (2017) found from an ensemble of inversions using surface and satellite observations that North American emissions had been flat for 2000-2012 and that without an inverse model short-term trends can appear to be present in the GOSAT data because of interannual transport variability, choice of background, and the seasonal sampling bias of GOSAT. Sheng et al. (2018a) analyzed 2010-2016 GOSAT enhancements over North America and found a $2.5 \pm 1.4\% \text{ a}^{-1}$ increase over the US driven by oil/gas production and livestock emissions in the Midwest. They found no significant trend over Canada (but large year-to-year variation driven by wetlands) and a $0.8\text{-}1.7\% \text{ a}^{-1}$ decrease in Mexican emissions driven by a decrease in livestock. Using 2006-2015 surface and aircraft observations over the US and Canada, Lan et al. (2019) found a $0.7 \pm 0.3\% \text{ a}^{-1}$ increase in total US emissions and a $3.4 \pm 1.4\% \text{ a}^{-1}$ increase in oil/gas emissions based on stations in Oklahoma, North Dakota, and Texas. They also reported an increase in the ethane:methane emission ratio which could lead to an overestimate of the oil/gas methane trend as inferred from the ethane trend.

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Our inverse analysis of the 2010-2015 GOSAT data over North America optimizes both mean emissions and their long-term trends at high resolution (up to 50 km). The inversion uses dynamic boundary conditions from a consistent global inversion of the 2010-2015 GOSAT data previously reported by Maasakkers et al. (2019). We use an analytical solution to the Bayesian inverse optimization problem (Jacob et al., 2016), which provides closed-form characterization of the information content of the solution, and also enables us to perform a range of sensitivity inversions (inversion ensemble) at no added computational cost. We relate the results from our inversion ensemble to the EPA GHGI emissions in order to inform knowledge of US emissions, their trends, and the contributions from different sectors.

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2 Data and methods

We perform a continental-scale inversion of 2010-2015 GOSAT methane data from the University of Leicester proxy CH_4 retrieval (Parker et al., 2015; ESA CCI GHG project team, 2018). We use the individual GOSAT observations to optimize a state vector of mean methane emissions and linear emission trends at spatial resolution of up to $0.5^\circ \times 0.625^\circ$ ($\approx 50 \times 50 \text{ km}^2$). The forward model for the inversion is the GEOS-Chem chemical transport model (www.geos-chem.org) applied in nested mode over North America with a spatial resolution of $0.5^\circ \times 0.625^\circ$. The main results presented here will be from a base inversion with specifications given below. In addition to this base inversion, we conducted an ensemble of 9 inversions in which we explored the sensitivity of the results to different assumptions. Specifications for these sensitivity inversions are given below and summarized in Section 2.5. We The posterior error covariance matrix from the inversion underestimates the actual uncertainty in the results because of the assumption of fully random observational errors (Jacob et al., 2016). Therefore

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we use the range of results from the inversion ensemble as a ~~range of uncertainty in the results from the base inversion~~better measure of uncertainty (Heald et al., 2004).

2.1 GOSAT observations

- 5 The GOSAT satellite has been observing dry column methane mixing ratios in the SWIR using the TANSO-FTS instrument since April 2009 (Butz et al., 2011). GOSAT in its default mode observes circular pixels of 10 km diameter at 13:00 local time, separated by ~ 250 km along-track and cross-track, repeating observation on the same track every 3 days. Additional locations ~~can~~are be observed using ~~the~~a target mode. GOSAT methane retrievals have a 24% success rate, limited mainly by cloud cover. Observations have a precision of 13 ppb and relative bias of 2 ppb compared to the Total Carbon Column Observing
- 10 Network (Buchwitz et al., 2015). There ~~is~~has been no significant spectral degradation of the observations over time (Kuze et al., 2016). Figure 1 shows the 156,110 retrievals over land used to optimize emissions in this study (Parker et al., 2015). Each retrieval comes with an estimated retrieval error (11 ppb on average). We use observations over land from January 2010 to December 2015, excluding data above 60°N for which model errors are large (Maasakkers et al., 2019). Most observations (95,365) are over the Contiguous United States (CONUS). The data are spatially sparse but this reflects the observing strategy
- 15 of repeated measurements at the same locations in the default mode. Thus most observation locations in Figure 1 have a large number of data points to inform temporal variability and trends (Sheng et al., 2018a).

2.2 Prior estimates

- Table 1 summarizes our prior emissions estimates and Figure 2 shows their spatial distributions for the major sectors. For all
- 20 US anthropogenic emissions including offshore as reported to the UNFCCC, we use the spatially disaggregated (gridded) version of the EPA GHGI (EPA, 2016) for 2012 from Maasakkers et al. (2016), with improved spatial allocation of emissions and detailed separation of source sectors compared to EDGAR. For oil/gas emissions in Mexico and Canada including offshore, we use gridded versions of the Instituto Mexicano del Petróleo (IMP) inventory for 2010 (IMP, 2012) and the ICF International inventory for 2013 (ICF, 2015), respectively, as reported by Sheng et al. (2017). IMP (2012) oil/gas emissions for Mexico match
- 25 the values reported by Mexico to UNFCCC, while ICF (2015) oil/gas emissions for Canada are 43% higher than the value reported by Canada (UNFCCC, 2019). The ICF inventory was used as the basis for the Sheng et al. (2017) gridded emission inventory as it provides a detailed breakdown of sources using methodology similar to the EPA GHGI. For other anthropogenic emissions in Canada/Mexico and other countries in the domain, we use the EDGAR v4.3.2 global emission inventory for 2012 (edgar.jrc.ec.europa.eu [2017]). We separate the general ‘fuel exploitation’ sector reported by EDGAR v4.3.2 into oil/gas and
- 30 coal components by using additional information embedded in the inventory (Greet Maenhout, personal communication). This allows us to use EDGAR coal emissions for Canada and Mexico. The emissions as given by EDGAR v4.3.2 are aseasonal. For manure management and rice cultivation we apply seasonality as prescribed by Maasakkers et al. (2016) and Zhang et al.

(2016) respectively. Other anthropogenic emissions remain aseasonal.

Natural emissions are dominated by wetlands for which we use mean monthly emissions from the WetCHARTS v1.0 extended ensemble with $0.5^\circ \times 0.5^\circ$ spatial resolution (Bloom et al., 2017). The ensemble parameters consist of: three global scaling factors (global emissions of 124.5, 166, or 207.5 Tg a⁻¹); three temperature dependencies (1, 2, or 3); and two landcover databases that are combined with precipitation data to estimate wetland extent (GLWD from Lehner and Döll (2004) or GlobCover from Bontemps et al. (2011)). Total wetland emissions vary month to month and interannually as driven by temperature and inundation extent (Bloom et al., 2017). Because the WetCHARTS ensemble exhibits considerable but uncertain year-to-year variability, we also perform a sensitivity inversion without prior interannual variability in wetland and other emissions. Daily open fire emissions are from the Quick Fire Emissions Dataset (QFED) (Darmenov and da Silva, 2013) and termite emissions are from Fung et al. (1991) with a global total of 12 Tg a⁻¹. We use geological seepage emissions compiled from literature on both point sources (Etiope, 2015; Kvenvolden and Rogers, 2005) and areal seepage (Kvenvolden and Rogers, 2005; Etiope and Klusman, 2010) as described in Maasakkers et al. (2019) with a global total of 5 Tg a⁻¹, under the 5.4 Tg a⁻¹ maximum proposed for pre-industrial times by Hmiel et al. (2020) based on ice core measurements.

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2.3 Forward model

We use the nested version of the GEOS-Chem chemical transport model v11-01 at $0.5^\circ \times 0.625^\circ$ resolution over North America as forward model for the inversion. Earlier versions of this model for methane were described by Wecht et al. (2014) and Turner et al. (2015). The model is driven with MERRA-2 meteorological fields (Bosilovich et al., 2016) from the NASA Global Modeling and Assimilation Office (GMAO). Methane loss from reaction with OH and Cl radicals, soil uptake, and stratospheric oxidation are described in Maasakkers et al. (2019). The simulation is initialized in January 2009 with concentration fields from Turner et al. (2015). Three-hourly boundary conditions at the edges of the nested domain are from the $4^\circ \times 5^\circ$ posterior model simulation of Maasakkers et al. (2019), which provides an unbiased fit to the global GOSAT data. That posterior simulation includes some information from GOSAT data over the North America domain, which were used (along with the more abundant data outside that domain) in the global inversion; but the main consideration here is to avoid bias in boundary conditions that would otherwise affect the North American inversion. Methane chemical and soil sinks are not optimized in our inversion because they are very slow compared to the time scale for ventilation of the North American domain.

Following Maasakkers et al. (2019), we correct the GEOS-Chem simulation of GOSAT columns for a latitudinally and seasonally variable background bias likely caused by the extratropical stratosphere (Bader et al., 2016; Saad et al., 2016; Stanevich, 2018). The bias is common in atmospheric models and caused by excessive meridional transport in the stratosphere (Patra et al., 2011) and in particular in the seasonal polar vortices (Zhang et al., 2020b). The latitudinal correction term ξ (ppb)

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Table 1. Methane emissions used as prior 2010-2015 estimates^a.

Source (Tg a ⁻¹)	CONUS	Canada	Mexico	Other ^b
Natural	15.7	15.3	1.4	3.8
Wetlands	14.2	14.4	1.0	3.4
Open fires	0.5	0.3	0.2	0.1
Termites	0.6	0.3	0.1	0.2
Geological seeps	0.5	0.3	0.1	$\theta < 0.1$
Anthropogenic	28.7	4.5	5.3	5.1
Livestock	9.2	1.0	2.5	1.9
Enteric Fermentation	6.7	0.8	2.2	1.9
Manure Management	2.5	0.2	0.3	0.1
Oil and Natural Gas	9.1	2.4	1.5	1.2
Gas production	4.4	1.2	0.1	
Oil production	2.3	0.5	1.2	
Gas Transmission	1.1	0.3	$\theta < 0.1$	
Gas Processing	0.9	0.3	0.1	
Gas Distribution	0.5	$\theta < 0.1$	$\theta < 0.1$	
Landfills	5.8	0.7	0.4	0.6
Coal Mining	2.9	0.1	$\theta < 0.1$	$\theta < 0.1$
Wastewater	0.7	0.2	0.7	0.7
Rice Cultivation	0.5	$\theta < 0.1$	$\theta < 0.1$	0.2
Other Anthropogenic ^c	0.5	0.1	0.2	0.4
Total Source	44.5	19.8	6.7	8.9

^a CONUS anthropogenic emissions are from the EPA GHGI for 2012 as spatially disaggregated by Maasakkers et al. (2016). Oil/gas emissions from Canada (2013) and Mexico (2010) are from ICF (2015) and IMP (2012), respectively, spatially disaggregated by Sheng et al. (2017). Other anthropogenic emissions are from EDGAR v4.3.2 for 2012 (edgar.jrc.ec.europa.eu [2017]). Wetlands and open fire emissions are mean values for 2010-2015 from the WetCHARTS ensemble (Bloom et al., 2017) and QFED (Darmenov and da Silva, 2013); Termite emissions are from Fung et al. (1991). Seepage emissions are as described in Maasakkers et al. (2019). The soil sink is 3.6 Tg a⁻¹ for the inversion domain (Fung et al., 1991) and is not optimized in the inversion. All values in the table are rounded to one decimal.

^b within the inversion domain shown in Figure 1 (10-70N°, 140-40W°) containing parts of Central and South America.

^c including fossil fuel combustion, industrial processes, agricultural field burning, and composting.

follows a quadratic form as in Turner et al. (2015):

$$\xi = (4.0\theta^2 - 1.3\theta) \times 10^{-3} - 5 \quad (1)$$

with θ the latitude in degrees. The seasonal bias is corrected over rolling 8° latitudinal bands. A sensitivity inversion without the seasonal bias correction is performed as part of the inversion ensemble.

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2.4 State vector for the inversion and error covariances

Although we could technically carry out the inversion of the GOSAT data at the $0.5^\circ \times 0.625^\circ$ resolution of the GEOS-Chem simulation, the data do not have sufficient information to constrain emissions on that grid and doing so would incur large smoothing error (Wecht et al., 2014). We use instead a 600-element Gaussian mixture model (GMM) as described by Turner and Jacob (2015) to optimally define the emission patterns that can be usefully constrained by the inversion. Each of the 600 Gaussian functions in the GMM is defined by an emission amplitude, mean location, and spread (standard deviation). These parameters are optimized using a similarity vector on the $0.5^\circ \times 0.625^\circ$ grid that takes into account latitude, longitude, and the prior patterns of different source sectors. The state vector \mathbf{x} for the inversion with dimension $n = 2 \times 600$ consists of scaling factors adjusting the amplitudes of the Gaussians in the GMM and their 2010-2015 linear trends. This approach allows for effective aggregation of regions with weak or homogeneous emissions while preserving high resolution for concentrated emissions. Each $0.5^\circ \times 0.625^\circ$ grid cell is represented by a unique combination of the Gaussians, so that the optimization of \mathbf{x} can be mapped to the $0.5^\circ \times 0.625^\circ$ grid. For more details see Turner and Jacob (2015).

Prior emission error variances are defined for each Gaussian on the basis of its spatial distribution and the contributions from different sectors. Emission errors for individual anthropogenic sectors are estimated using the error curves from Maasakkers et al. (2016). The error standard deviation σ for a given source sector and Gaussian is given by:

$$\sigma = (\alpha_0 \exp(-k_\alpha(L - L_0)) + \alpha_N) E \quad (2)$$

Where α_0 , k_α , and α_N are source sector specific error coefficients from Maasakkers et al. (2016), L is the effective spatial resolution (length scale) of the Gaussian defined by the number of $0.5^\circ \times 0.625^\circ$ grid cells it represents, $L_0 = 0.1^\circ$ is the native resolution of the prior inventory, and E is the sum of emissions from the source sector within the Gaussian (sum of emissions from $0.5^\circ \times 0.625^\circ$ grid cells weighted by their contributions to the Gaussian). Maasakkers et al. (2016) also include a displacement error related to uncertainty in source location but this error is negligible at our resolution. For wetland emissions, we use the standard deviation in monthly estimates of the 18 WetCHARTS v1.0 extended ensemble members averaged over the Gaussian; the resulting error standard deviation is 78% on average. For the other natural emissions we assume 100% error at the $0.5^\circ \times 0.625^\circ$ model resolution.

The error variances for all sectors contributing to a given Gaussian are added in quadrature to obtain the corresponding diagonal element of the prior error covariance matrix \mathbf{S}_A . Error variances for a given Gaussian are capped at 50% in the base inversion and we also perform a sensitivity inversion without this cap. The mean relative error standard deviation is 37% in the base inversion. The 50% cap mainly affects Gaussians dominated by wetland emissions. For the 2010-2015 emission trends associated with each Gaussian, the prior estimate is set to zero and the prior error standard deviation is a 5% change per year, in line with uncertainties in trend estimates for North America (Turner et al., 2016; Bruhwiler et al., 2017; Sheng et al., 2018a; Lan et al., 2019). We also perform sensitivity inversions with changes of 2.5% and 10% per year as prior error standard deviation. Off-diagonal elements of \mathbf{S}_A are assumed to be zero because Maasakkers et al. (2016) found no spatial error correlation for the gridded EPA inventory; this may be an underestimate for wetland emissions (Bloom et al., 2017).

Our calculation of \mathbf{S}_A leads to different error variances for each grid cell. To assess the impact of that choice, we also perform a sensitivity inversion using the mean error variance for all the Gaussians. The base inversion assumes normal errors, but we also perform a sensitivity inversion assuming log-normal emission errors following the Levenberg-Marquardt method as described in Maasakkers et al. (2019).

We use the residual error method (Heald et al., 2004) to construct the diagonal of the observational error covariance matrix \mathbf{S}_O . The mean 2010-2015 difference between GOSAT and the prior model (before seasonal correction) for each $0.5^\circ \times 0.625^\circ$ grid cell is assumed to be due to errors in emissions, to be corrected by the inversion. After subtracting this mean difference, the residual standard deviation is taken as estimate of the observational error standard deviation including contributions from instrument, representation, and forward model errors. If this estimate is less than the reported instrument error standard deviation (Parker et al., 2015), we use the latter instead (17% of observations). If it is less than 10 ppb we reset it to 10 ppb (6% of observations). The resulting average observational error standard deviation is 14 ppb. Off-diagonal terms of \mathbf{S}_O are assumed to be zero for lack of better information, but in fact some transport error correlation would be expected in the forward model. We account for this error correlation with a regularization term γ in the inversion (Section 2.5).

2.5 Inversion procedure

We perform an analytical inversion minimizing the Bayesian cost function $J(\mathbf{x})$ assuming normal errors (Rodgers, 2000):

$$J(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_A)^T \mathbf{S}_A^{-1} (\mathbf{x} - \mathbf{x}_A) + \gamma (\mathbf{y} - F(\mathbf{x}))^T \mathbf{S}_O^{-1} (\mathbf{y} - F(\mathbf{x})) \quad (3)$$

where \mathbf{x} is the state vector to be optimized, consisting of 600 Gaussians for which we optimize both scaling factors for mean emissions and absolute linear emission trends, for a total of 1200 state vector elements; \mathbf{x}_A is the prior state vector; \mathbf{S}_A is the prior error covariance matrix (Section 2.4); \mathbf{S}_O is the observational error covariance matrix (Section 2.4); and γ is a regularization factor to account for the lack of non-diagonal terms in \mathbf{S}_O and hence prevent overfitting. γ plays a similar role as the regularization parameter in Tikhonov methods (Brasseur and Jacob, 2017) and reflects our inability to precisely quantify

error statistics in the Bayesian method. Here we find that $\gamma = 0.5$ provides the best balance of fitting the prior and observational terms in the cost function, following the L-curve approach of Hansen (1999). The value is higher than $\gamma = 0.05$ used in the global inversion of Maasakkers et al. (2019) at $4^\circ \times 5^\circ$ resolution because here we have a smaller number of observations per state vector element. We also conduct sensitivity inversions with $\gamma = 0.1$ and $\gamma = 1$.

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The GEOS-Chem forward model ($\mathbf{y} = F(\mathbf{x})$) as implemented here is strictly linear in its relationship between methane column concentrations (\mathbf{y}) and the state vector of emissions (\mathbf{x}). It can be expressed as $F(\mathbf{x}) = \mathbf{K}\mathbf{x} + \mathbf{c}$ where $\mathbf{K} = \partial\mathbf{y}/\partial\mathbf{x}$ is the Jacobian matrix and \mathbf{c} an initialization constant. This allows the optimal posterior solution $\hat{\mathbf{x}}$ which minimizes the cost function $J(\mathbf{x})$ to be obtained analytically as:

$$10 \quad \hat{\mathbf{x}} = \mathbf{x}_A + \mathbf{S}_A \mathbf{K}^T \left(\mathbf{K} \mathbf{S}_A \mathbf{K}^T + \frac{\mathbf{S}_O}{\gamma} \right)^{-1} (\mathbf{y} - \mathbf{K} \mathbf{x}_A) \quad (4)$$

with posterior error correlation matrix $\hat{\mathbf{S}}$:

$$\hat{\mathbf{S}} = (\gamma \mathbf{K}^T \mathbf{S}_O^{-1} \mathbf{K} + \mathbf{S}_A^{-1})^{-1} \quad (5)$$

The information content from the inversion can then be obtained from the averaging kernel matrix ($\mathbf{A} = \partial\hat{\mathbf{x}}/\partial\mathbf{x}$) which gives the sensitivity of the solution to the true state:

$$15 \quad \mathbf{A} = \mathbf{I} - \hat{\mathbf{S}} \mathbf{S}_A^{-1} \quad (6)$$

The trace of \mathbf{A} gives the degrees of freedom for signal (DOFS), which measures the number of independent pieces of information on the state vector that can be obtained from the inversion. The diagonal elements of \mathbf{A} (averaging kernel sensitivities) measure the degree to which the inversion can constrain the true values of the corresponding state vector elements (1 = perfectly, 0 = not at all). We will use these measures of information in our presentation of results.

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Analytical solution to the inverse problem requires explicit construction of the Jacobian matrix. We perform this construction column by column by perturbing individually the 1200 elements of the state vector and conducting the corresponding GEOS-Chem simulations for the 2010-2015 observation record. This is readily done as a massively parallel calculation. Aside from enabling closed-form characterization of the information content from the inversion, a major advantage of the analytical

25 solution once the Jacobian matrix has been constructed is that the sensitivity of the solution to various assumptions and choices made in the inversion approach can be immediately obtained. In addition to our base inversion, we generate in this manner an ensemble of nine sensitivity inversions introduced in the text above, and for which the ensemble of solutions gives a better measure of posterior error than can be obtained from $\hat{\mathbf{S}}$ (Heald et al., 2004). To summarize, these sensitivity inversions include:

30 (1) Using a prior estimate with no interannual variability (2012 values) for wetland and biomass burning emissions, and for the soil sink; (2) Not using a seasonal correction to the GOSAT - model mismatch; (3) Using emission error variances without

the 50% cap; (4) Using the average emission and absolute trend error variances (37% and $2.3 \text{ Mg a}^{-2} \text{ km}^{-2}$, respectively) for each Gaussian; (5,6) Assuming error prior standard deviations for the 2010-2015 trend of 2.5% and 10% annual change of emissions (instead of 5%); (7) Assuming log-normal prior emission errors; (8,9) Using regularization factors for the cost function $\gamma = 0.1$ and $\gamma = 1$ (instead of $\gamma = 0.5$).

5

3 Results and Discussion

Figure 3 shows mean prior and posterior emissions for 2010-2015, the ratio between the two, and the inversion's averaging kernel sensitivities (Equation 6). The averaging kernel sensitivities identify regions where the GOSAT observations provide significant information on emissions. These are regions with a high density of observations and/or high absolute uncertainties on the prior emissions. For example, we achieve good constraints on emissions in central Canada, much of the eastern and central US, California, and southeastern Mexico. Other regions receive little information from the observations, which explains a lack of departure from the prior estimate.

The posterior emissions when implemented in GEOS-Chem reduce the mean squared difference with GOSAT observations by 3.5%. ~~Overall changes are~~ This overall reduction in error is small because random errors in individual observations are large and because the background is already captured well in the prior simulation through the optimized boundary conditions. The main improvements are found over areas where the averaging kernels are large (Figure 3). For data with averaging kernel sensitivities greater than 0.1, the mean squared difference is reduced by 6.1% and the correlation increases from 0.62 to 0.64. We independently evaluated the posterior estimate by comparison to in situ methane concentrations from surface sites reported in the GLOBALVIEWplus CH₄ ObsPack v1.0 data product compiled by the NOAA Global Monitoring Laboratory (Cooperative Global Atmospheric Data Integration Project, 2019). Compared to the prior simulation (Reduced major axis (RMA) slope: 0.69, $r^2 = 0.39$), the posterior simulation (RMA slope: 0.69, $r^2 = 0.45$) does not degrade the comparison with these data and improves the correlation. The spatial coefficient of determination between the time-averaged GEOS-Chem and NOAA data increases from $r^2 = 0.58$ with the prior emissions to $r^2 = 0.81$ with the posterior emissions, representing an improvement in our ability to fit observed patterns.

3.1 Mean 2010-2015 emissions

Although the inversion yields little change in total emissions for the continental domain, there are large regional changes as shown in Figure 3. We find higher emissions over the south-central and eastern US, and lower emissions in California compared to the gridded EPA inventory. The WetCHARTS inventory overestimates wetland emissions including along the Gulf and east coasts of the US, the upper Midwest, and Canada. Emissions in eastern Mexico are higher than inferred from the IMP (2012) inventory. The inversion also shows large relative increases from oil production off the Louisiana coast and from

wetlands/livestock in western Montana but the associated emissions are low. The large-scale correction patterns revealed by the inversion are similar to those of the coarse ($4^\circ \times 5^\circ$) global inversion reported by Maasakkers et al. (2019), which used the same prior estimates, but we have much more detail here allowed by the higher resolution of the inversion.

5 Figure 4 shows the attribution of the inversion results to individual source sectors for CONUS, Canada, and Mexico. This attribution was made by applying the correction factors to the sectoral emissions in each grid cell, assuming that the relative contributions of individual sectors to emissions in that grid cell is correct in the prior emission inventory (this does not assume that the total prior distribution of sectoral emissions is correct). Vertical bars show the range of results from the inversion ensemble. ~~Narrow ranges may~~ A narrow uncertainty range does not necessarily reflect confidence in the inversion results ~~but the~~
10 ~~inability of the inversion to inform emissions from a particular sector when it is small.~~ For small source sectors, it may also be due to insufficient information from the observations so that the optimization is unable to depart from the prior estimate. This can be determined using the averaging kernel sensitivities, as will be done below for the US (see Table 2).

The largest decrease is for US wetland emissions, mostly contributed by the Gulf and east coasts (Figure 2). Such an over-
15 estimate in the mean of the WetCHARTS wetland inventory ensemble was previously identified in an inversion of aircraft observations over the Southeast US (Sheng et al., 2018b). It may be related to the low organic carbon content of the soil, the difficulty of distinguishing freshwater and saltwater wetlands, uncertainties in anaerobic CH_4 : CO_2 respiration rates, and the accounting of partial wetland land-cover areas (Holmquist et al., 2018; Lehner and Döll, 2004; Bloom et al., 2017). We also find an overestimate of wetland emissions in (mainly eastern) Canada. The large uncertainty range is driven by the inversion en-
20 semble member without seasonal correction. Based on the root mean square error and spatial correlation, our inversion results are most consistent with the WetCHARTS ensemble members that use GlobCover wetland extent, a $q_{10} = 2$ value for the factor increase in the CH_4 to CO_2 emission ratio per 10 K temperature increase (a critical quantity for determining the sensitivity of wetland CH_4 production to temperature (Yvon-Durocher et al., 2014; Bloom et al., 2016)), and global scaling at the low end or middle of the range (global wetlands emission range 125-166 Tg a^{-1}). A value of $q_{10} = 2$ is approximately equivalent with the
25 average CH_4 to CO_2 temperature sensitivity reported by Yvon-Durocher et al. (2014) based on meta-analyses, which indicates that anaerobic CH_4 respiration is substantially more sensitive to temperature relative to overall CO_2 respiration rates. Sheng et al. (2018b) also found their inversion results to be most consistent with GlobCover but also favored no CH_4 : CO_2 temperature dependence ($q_{10} = 1$). Their observations were much more limited in space and time (August-September 2013).

30 Figure 4 also shows some significant sectoral corrections for anthropogenic emissions in the US and Mexico. Over Mexico higher livestock (+13 (5-24)%) and oil/gas emissions (+22 (-24-42)%). Uncertainty ranges in correcting individual sectors are large for Mexico because of the extensive spatial overlap between sectors (Figure 2). Most of the oil/gas correction is for coastal/offshore oil production (Figure 3). We also find 56 (31-120) % higher emissions over Mexico City, which is optimized by a single Gaussian covering five grid cells. The difference is attributed to wastewater based on the EDGAR spatial patterns.
35 Compared to EDGAR v4.3.2 a recent gridded inventory for Mexico (Scarpelli et al., 2020) and the Mexico City Secretariat

of Environment (~~SEDEMA~~) (SEDEMA, 2018) air quality emission inventory (~~SEDEMA, 2018~~) ~~predicts~~ predict lower emissions from wastewater (68 versus 259 Gg a⁻¹ in the SEDEMA inventory) but much higher landfill emissions (222 versus 1 Gg a⁻¹) indicating that our higher emission estimate may be related to landfill emissions being misallocated in EDGAR v4.3.2.

5 Inversion results for the US mapped on to the detailed source sectors and subsectors from the gridded EPA inventory (Maasakkers et al., 2016) are given in Table 2. The ~~table~~ Table also includes the averaging kernel sensitivity ~~a_i~~ $a_{i,i}$ from the inversion for each sector and subsector i , which we estimate by summing emissions from sector/subsector i for all $0.5^\circ \times 0.625^\circ$ grid cells over the CONUS into one state vector element using a summation matrix (Calisesi et al., 2005; Maasakkers et al., 2019). The ~~one-dimensional~~ summation matrix (~~$\mathbf{W}_i = (w_{i,k})\mathbf{W}$~~) weighs the relative contribution $w_{i,k}$ of sector/subsec-

10 tor i to the total emission in Gaussian k in the prior inventory:

$$\underline{a_i} \underline{\mathbf{A}_{sub}} = \underline{\mathbf{W}_i} \underline{\mathbf{W}} \underline{\mathbf{A}} \underline{\mathbf{W}_i^*} \underline{\mathbf{W}^*} \quad (7)$$

$$\underline{\hat{\mathbf{S}}_{sub}} = \underline{\mathbf{W}} \underline{\hat{\mathbf{S}}} \underline{\mathbf{W}^T} \quad (8)$$

Here ~~$\mathbf{W}_i^* = \mathbf{W}_i^T (\mathbf{W}_i \mathbf{W}_i^T)^{-1} \mathbf{W}^* = \mathbf{W}^T (\mathbf{W} \mathbf{W}^T)^{-1}$~~ is the generalized pseudo-inverse of ~~\mathbf{W}_i~~ , and ~~\mathbf{A} is \mathbf{W}~~ , and ~~\mathbf{A}_{sub}~~ (with diagonal elements $a_{i,i}$) and ~~$\hat{\mathbf{S}}_{sub}$~~ are the averaging kernel matrix (~~Equation 6~~), ~~$a_i = 1$~~ and posterior error covariance matrix mapped to the different subsectors. $a_{i,i} = 1$ means that the inversion can fully constrain the national total for that emission category, independent of the prior estimate, while ~~$a_i = 0$~~ $a_{i,i} = 0$ means that the inversion provides no information and the estimate cannot depart from the prior. The off-diagonal elements of $\hat{\mathbf{S}}_{sub}$ measure the error correlation in the posterior solution for different subsectors, and this is important to diagnose whether we can optimize different subsectors independently. The diagonal elements of ~~$\hat{\mathbf{S}}_{sub}$~~ estimate the error variance in the posterior solution for individual subsectors but that estimate is too small because it assumes that the observations are independent and identically distributed (IID condition) (Brasseur and Jacob, 2017). We prefer to estimate the error in the posterior solution from the results of the inversion ensemble, as shown in Table 2.

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25 The averaging kernel sensitivities for individual sectors/subsectors in Table 2 vary based on the uncertainty of the prior emission estimates and the GOSAT observation density in the regions of emissions. Posterior wetland emissions (~~$a_i = 0.68$~~ are 68% ~~$a_{i,i} = 0.70$~~ are 70% informed by the observations (~~32~~ 30% by the prior) because the prior uncertainty is large. ~~Total US anthropogenic emissions are 60~~ We also calculate $a_{i,i}$ for the sum of US anthropogenic emission categories and find emissions are 53% informed by the observations, with less information for individual sectors/subsectors. Emissions from oil/gas produc-

30 tion are particularly well informed (~~45-54~~ 28-52%) because they are large and have relatively little spatial overlap with other sectors.

Our posterior estimate for the mean 2010-2015 CONUS anthropogenic source is 30.6 (29.4-31.3) Tg a⁻¹, where the best estimate is from the base inversion and the range is from the inversion ensemble. The 2012 emission total from the EPA GHGI (EPA, 2016) used as prior estimate in our inversion is 28.7 Tg a⁻¹, with an uncertainty range 26.4-36.2. We find limited posterior error correlation ($r = 0.33$) between the posterior anthropogenic and natural emission totals. Examining the contributions from different sectors, our best posterior estimates for landfills and livestock are within 5% of the GHGI, and coal emissions are 6% higher. Oil/gas emissions total 11.1 Tg a⁻¹ in our base inversion, 22 (12-32)% higher than the GHGI, and driven by oil/gas production as seen for example in Texas, Oklahoma, and offshore in the Gulf of Mexico. Our national estimates for the emissions from oil and gas production are 3.1 (2.7-3.6) and 5.4 (4.9-5.9) Tg a⁻¹, respectively, as compared to 2.3 and 4.4 Tg a⁻¹ in the GHGI. The posterior error covariance between wetland emissions and both oil production ($r = 0.02$) and gas production ($r = 0.04$) are low, showing that this increase is independent of the large decrease in wetland emissions.

Our scaling factors to the EPA GHGI are for the 2012 emissions as reported by EPA (2016) and used in the inversion as prior estimates. More recently, EPA (2020) updated its methodology for estimating emissions and applied it to a reanalysis of emissions from previous years including 2012. Changes for 2012 emissions are important for some oil/gas subsectors, as shown in Figure 5. Gas production emissions in 2012 are lower by 19% in the updated GHGI because of a downward correction to emissions from gathering and boosting emissions stations. Oil production emissions in 2012 are 30% lower ~~than previously reported, caused by a correction for wells that were previously double-counted~~ in the updated EPA GHGI because of previous faulty double-counting of wells. Our correction factor from the inversion increases oil production emissions by a factor 1.9 (1.7-2.3) and natural gas production emissions by a factor 1.5 (1.4-1.6) relative to the updated 2012 GHGI from EPA (2020). ~~Similarly, emissions from~~ The updated GHGI emissions from natural gas processing in ~~the updated GHGI 2012~~ are 55% lower than previously reported, but our inversion finds them to be higher. Our correction factor from the inversion increases gas processing emissions by a factor of 2.9 (2.6-3.1) relative to the updated GHGI.

3.2 2010-2015 Emission trends

Figure 6 shows linear emission trends for 2010-2015 optimized by the base inversion (top left panel) and the sensitivity inversion including no interannual variability in prior estimates for wetlands and open fires (top right panel). The base inversion shows a trend of increasing emissions from US wetlands, but this is relative to the prior WetCHARTS estimate of interannual variability of wetland emissions which vary from 31 Tg in 2015 to 36 Tg in 2010. Due to the sparsity of independent constraints, the WetCHARTS inter-annual variations have not been extensively evaluated (Bloom et al., 2017), therefore we have little confidence in these year-to-year emission changes. The sensitivity inversion including no prior interannual variability for wetlands shows no large trends in US wetland emissions. Both inversions show similar results for the emission trends in anthropogenic source regions. We find an increase in total anthropogenic CONUS emissions of 0.14 Tg a⁻¹ a⁻¹ (0.4 % a⁻¹) over the 2010-2015 period. This anthropogenic trend is much lower than the $2.8 \pm 0.3\%$ a⁻¹ increase reported for 2010-2014 by Turner

Table 2. Mean 2010-2015 methane emissions in the contiguous US (CONUS)

Source (Tg a ⁻¹)	Prior estimate ^a	Posterior estimate ^b	Sensitivity ^c
Natural	15.7	11.8 (7.1-12.7)	0.62 <u>0.63</u>
Wetlands	14.2	10.2 (5.6-11.1)	0.68 <u>0.71</u>
Open fires	0.5	0.4 (0.4-0.5)	0.15 <u>0.13</u>
Termites	0.6	0.6 (0.6-0.6)	0.18 <u>0.02</u>
Geological seeps	0.5	0.5 (0.5-0.5)	0.16 <u>0.06</u>
Anthropogenic	28.7	30.6 (29.4-31.3)	0.60 <u>0.53</u>
Livestock			
Enteric Fermentation	6.7	6.9 (6.3-7.0)	0.32 <u>0.16</u>
Manure Management	2.5	2.5 (2.1-2.5)	0.31 <u>0.22</u>
Oil and Natural Gas			
Gas production	4.4	5.4 (4.9-5.9)	0.45 <u>0.28</u>
Oil production	2.3	3.1 (2.7-3.6)	0.54 <u>0.53</u>
Gas Transmission	1.1	1.1 (1.1-1.2)	0.17 <u>0.03</u>
Gas Processing	0.9	1.1 (1.0-1.2)	0.49 <u>0.41</u>
Gas Distribution	0.5	0.4 (0.4-0.4)	0.41 <u>0.35</u>
Landfills			
Municipal	5.2	5.0 (4.7-5.0)	0.39 <u>0.26</u>
Industrial	0.6	0.5 (0.5-0.5)	0.24 <u>0.13</u>
Coal Mining			
Underground	2.2	2.4 (2.3-2.5)	0.31 <u>0.22</u>
Surface	0.5	0.5 (0.4-0.5)	0.32 <u>0.30</u>
Abandoned	0.2	0.3 (0.3-0.3)	0.22 <u>0.10</u>
Wastewater			
Municipal	0.5	0.4 (0.4-0.4)	0.22 <u>0.09</u>
Industrial	0.2	0.2 (0.1-0.2)	0.19 <u>0.16</u>
Rice Cultivation	0.5	0.4 (0.3-0.5)	0.30 <u>0.28</u>
Other Anthropogenic ^d	0.5	0.4 (0.4-0.5)	0.34 <u>0.05</u>
Total Source	44.5	42.4 (37.0-42.9)	0.63 <u>0.64</u>

^aThe prior estimates include the 2012 EPA GHGI emissions (EPA, 2016) and the 2010-2015 mean of the WetCharts inventory ensemble for wetlands (Bloom et al., 2017).

^bPosterior estimates from our base inversion, with range from the inversion ensemble in parentheses.

^cSensitivity of the posterior estimate to the observations as diagnosed from the averaging kernel matrix, ranging from 0 (no sensitivity, posterior equal to prior) to 1 (full sensitivity, posterior solely determined by the observations). For example, a sensitivity of 0.64 means that 64% of the posterior estimate is constrained by the observations and 36% is constrained by the prior. Averaging kernel sensitivities can statistically be negative in case of error overlap with other sources. The small negative value here is insignificant.

^dIncluding fossil fuel combustion, industrial processes, agricultural field burning, and composting.

et al. (2016) and more in line with the 2006-2015 trend of $0.7 \pm 0.3 \text{ \% a}^{-1}$ in total US emissions estimated by Lan et al. (2019).

The GHGI (EPA, 2020) reports a $0.35 \text{ Tg a}^{-1} \text{ a}^{-1}$ decrease in anthropogenic US emissions from 2010 to 2015, at odds with our result. The decrease in the GHGI is mainly driven by decreasing emissions from landfills ($-0.10 \text{ Tg a}^{-1} \text{ a}^{-1}$) and coal mining ($-0.17 \text{ Tg a}^{-1} \text{ a}^{-1}$). We find small decreases in the western US that may be related to decreases in emissions from landfills or coal mines (Wyoming). On the national scale, however, ~~we find no evidence for the inversion~~ does not detect decreasing emissions from landfills or coal mining.

For total oil/gas emissions we find a US trend of $0.4 \text{ (0-1) \% a}^{-1}$ for 2010-2015, smaller than the $3.4 \pm 1.4 \text{ \% a}^{-1}$ increase reported by Lan et al. (2019) for 2006-2015. The discrepancy may be explained by the different time periods and the fact that the Lan et al. (2019) oil/gas trend is mainly determined by stations in Oklahoma, North Dakota, and Texas. Most of our increase is driven by the Marcellus Shale area in the Northeast US, amounting to $130 \text{ (20-190) Gg a}^{-1} \text{ a}^{-1}$. This area covering Pennsylvania, Ohio, and West Virginia has seen a large increase in natural gas production driven by unconventional drilling. Natural gas production in the area increased by a factor 7.8 between 2010 and 2015, contributing 22% of US natural gas production in 2015 (EIA, 2020a). If the $130 \text{ (20-190) Gg a}^{-1} \text{ a}^{-1}$ increase is entirely due to gas production, it would only amount to $15 \text{ (2-21) \% a}^{-1}$ relative to the mean posterior gas production emissions in the area (0.89 Tg a^{-1}) indicating that the leakage rate did decrease over the time period. The latest GHGI shows a national $60 \text{ Gg a}^{-1} \text{ a}^{-1}$ decrease in natural gas production emissions over the 2010-2015 period (EPA, 2020), mainly due to decreasing onshore production and exploration emissions and partly offset by increasing gathering and boosting emissions. For onshore production emissions, the GHGI primarily estimates emissions on the basis of the number of wells, rather than by production rate, and this may underestimate the trend in the Marcellus Shale as the number of wells only increased by 13% over 2010-2015 (EIA, 2020b) despite the large increase in production. The inversion suggests additional increases over production regions in Texas (Permian Basin) and Oklahoma. The Permian Basin has seen a large increase in production after 2015 (beyond the timespan of our inversion) and is currently the largest oil-producing Basin in the US (Zhang et al., 2020a).

GOSAT provides little information over Canada and Mexico when it comes to trends. There are signs that oil/gas production emissions in both Alberta (Canada) and offshore in the Gulf of Mexico are decreasing, and the latter may be driven by decreasing oil production (Zhang et al., 2019). In Canada, gas production has been stable while oil production from oil sands has increased, but the number of wells has decreased and efforts have been made to reduce emissions (Natural Resources Canada, 2020)

3.3 Comparison with other evaluations of the EPA inventory

A number of studies using atmospheric measurements over the US have previously compared their findings to the gridded version of the EPA GHGI as reported by Maasakkers et al. (2016) and used as prior estimate in our inversion. Based on an

upscaling of facility-level measurements and aircraft data, Alvarez et al. (2018) estimated 2015 US oil/gas emissions of 13 (11-15) Tg a⁻¹, consistent with our posterior estimate of 11.4 (10.3-12.2) Tg a⁻¹ for 2015 (posterior mean 2010-2015 emissions plus trend) and much higher than the 7.3 Tg a⁻¹ national total from the latest GHGI (EPA, 2020). Similar to our subsector attribution, Alvarez et al. (2018) find the largest difference with the GHGI for the production subsector (factor 2), which they attribute to the GHGI emissions not accounting for emissions from abnormal operating conditions. While Alvarez et al. (2018) did not distinguish between oil and gas production, our results point at a much larger relative discrepancy with the GHGI for oil production emissions than natural gas production emissions.

In an inversion of data from two tower networks and one aircraft campaign, Cui et al. (2019) found 2014-2016 California methane emissions to be 2.05 ± 0.26 Tg a⁻¹. Our posterior estimate of 2015 California emissions is 1.6 (0.8-1.7) Tg a⁻¹, representing a significant decrease from the prior estimate of 2.3 Tg a⁻¹. This is due to our large reduction of mean 2010-2015 emissions in the Los Angeles Basin. This reduction may be overestimated because of the coarseness of model CO₂ used in the proxy retrieval, underestimating CO₂ over Los Angeles (Turner et al., 2015). Using aircraft measurements, Ren et al. (2019) found 70% higher oil/gas production emissions in the Marcellus Shale (Pennsylvania/West Virginia) in 2015 compared to the 2012 gridded EPA inventory, which they attribute to an increase in production. We find a 2010-2015 increase in emissions for that region, as discussed above, but 2015 emissions are still only 22% higher than the GHGI. Based on surface observations in the Uintah Basin in Utah, Foster et al. (2017) found good agreement with basin-wide emissions from Karion et al. (2013), and found the gridded EPA inventory to be 45% lower after adjusting emissions based on 2015 production data. Most of the emissions in the Uintah Basin are concentrated in one of our grid cells and that cell is optimized individually in our inversion with good constraints (Fig 3), finding 2015 posterior emissions that are 37(12-66)% higher than the prior and no significant ~~linear~~ 2010-2015 trend.

Based on aircraft data, Plant et al. (2019) found a factor 2 higher anthropogenic urban methane emissions compared to the gridded EPA inventory over five cities on the US East Coast. We find no such difference but evaluating urban emissions along the East Coast is difficult because of overlap with large wetlands emissions that are themselves highly uncertain. It should be possible in principle to separate urban and wetland emissions on the basis of seasonality but we have little confidence in doing so with the GOSAT data because of the need for a seasonal correction to the model - GOSAT mismatch (Section 2.3) and the uncertainty in the seasonality of wetland emissions (Melton et al., 2013; Poulter et al., 2017).

4 Conclusions

We have used 2010-2015 methane column data from the GOSAT satellite instrument in a high-resolution inversion of methane emissions and their trends over North America during that period. The inversion for the contiguous US (CONUS) uses as prior estimate a gridded version of the EPA Inventory of U.S. Greenhouse Gas Emissions and Sinks (GHGI), so that results from the

inversion are directly relevant for evaluating the GHGI including the contributions from different sectors/subsectors to national methane emissions. We use a 600-member Gaussian mixture model (GMM) as state vector for the inversion that enables us to achieve high resolution ($0.5^\circ \times 0.625^\circ$) in concentrated source regions, and an analytic solution to the Bayesian inverse problem that includes full characterization of information content and facilitates the computation of an ensemble of sensitivity
5 inversions to estimate uncertainty.

We find a best estimate for mean US anthropogenic emissions in 2010-2015 of 30.6 Tg a^{-1} (range of $29.4\text{-}31.3 \text{ Tg a}^{-1}$ from the inversion ensemble), slightly higher than the EPA GHGI estimate of $28.7 \text{ (}26.4\text{-}36.2\text{) Tg a}^{-1}$. The difference is mainly from oil and gas production, which we find to be higher by 35% (19-59%) and 22% (11-33%) respectively compared to the GHGI.
10 The most recent version of the GHGI EPA (2020) revises emissions from oil/gas production and gas processing emissions downward, opposite from our results. Thus we find that the estimate of emissions from oil production by EPA (2020) is lower than our result by a factor of 2.

Our best estimate of CONUS wetland emissions is $10.2 \text{ (}5.6\text{-}11.1\text{) Tg a}^{-1}$, representing 24% of total CONUS methane
15 emissions. This is lower than the ensemble mean from the WetCHARTS inventory (14.2 Tg a^{-1}) used as prior estimate, and is consistent with previous studies pointing to overestimates in US wetland emissions. More work is needed to understand the underlying processes. We find a similar overestimate in wetland emissions over Eastern Canada. We estimate mean 2010-2015 anthropogenic emissions of $4.5 \text{ (}4.4\text{-}4.7\text{) Tg a}^{-1}$ for Canada and $6.1 \text{ (}5.5\text{-}6.3\text{) Tg a}^{-1}$ for Mexico. We find that oil/gas emissions in the IMP (2012) inventory reported to the UNFCCC are too low by 20%, mainly driven by oil production.

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We find from the inversion a 2010-2015 increase in US anthropogenic emissions of 0.14 Tg a^{-1} ($0.4 \% \text{ a}^{-1}$), much lower than previous GOSAT-based estimates but at odds with the latest EPA GHGI that reports a 0.35 Tg a^{-1} decrease in emissions over that period. Our increase appears to be largely driven by the rapid growth of unconventional oil/gas production in the eastern US.

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Author contributions. JDM and DJJ designed the study. JDM performed the analysis. JDM and MPS performed the simulations. JDM, DJJ, MPS, TRS, HN, JXS, YZ, XL, AAB, KWB, and JRW discussed the results. AAB provided the WetCHARTS emissions and supporting data. RJP provided the GOSAT data and supporting guidance. JDM and DJJ wrote the paper, and all authors provided input on the paper for revision before submission.

30 *Competing interests.* The authors declare that they have no conflict of interest.

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GOSAT Methane for 2010-2015

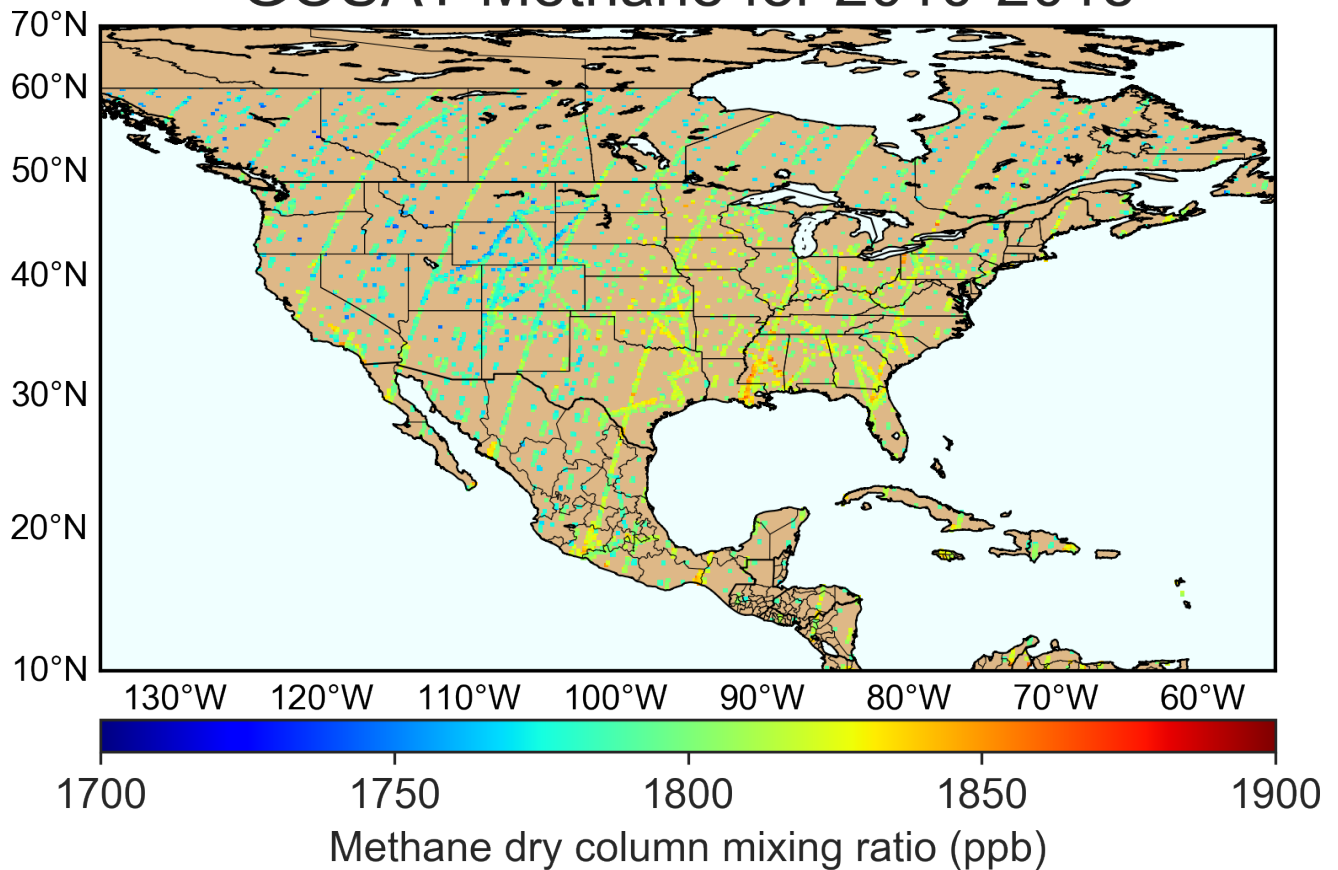


Figure 1. Average 2010-2015 methane dry column mixing ratios over North America observed by GOSAT. There are 156110 individual observations over land used in the inversion. The GOSAT data have 10km pixel resolution but we inflate them here to $0.3^{\circ} \times 0.3^{\circ}$ for visibility. GOSAT generally takes repeated observations of the same pixels so that most pixels shown here average a number of observations. The apparent north-south tracks that are GOSAT's default mode observations with a repeat cycle of 3 days, while the off-track data are target mode observations. GOSAT observations north of 60°N are excluded because of their seasonal limitation and uncertainty about the stratospheric correction.

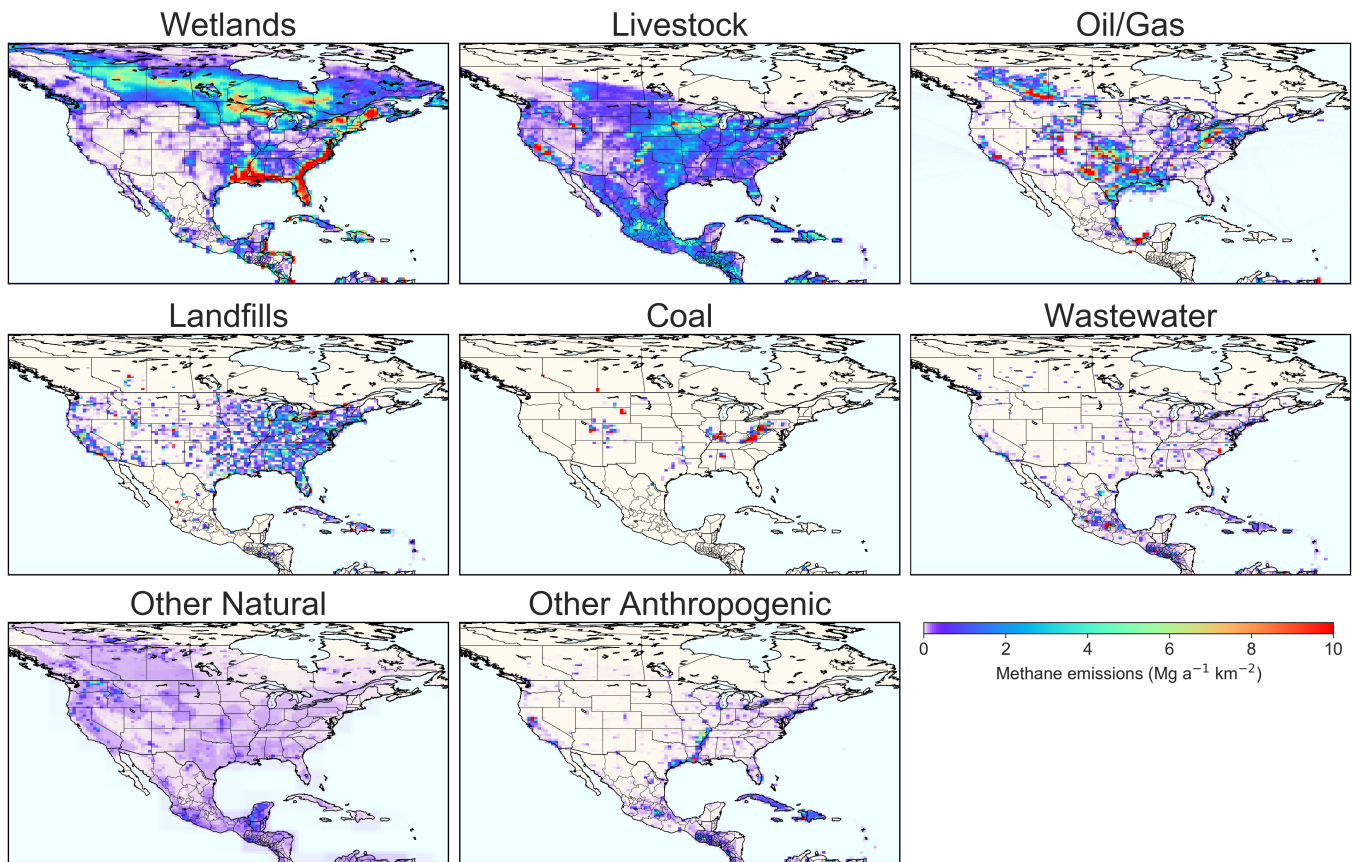


Figure 2. ~~Methane emissions taken as~~ Mean prior estimate estimates of methane emissions for 2010-2015. National totals, subsector breakdowns, and references are in Table 1.

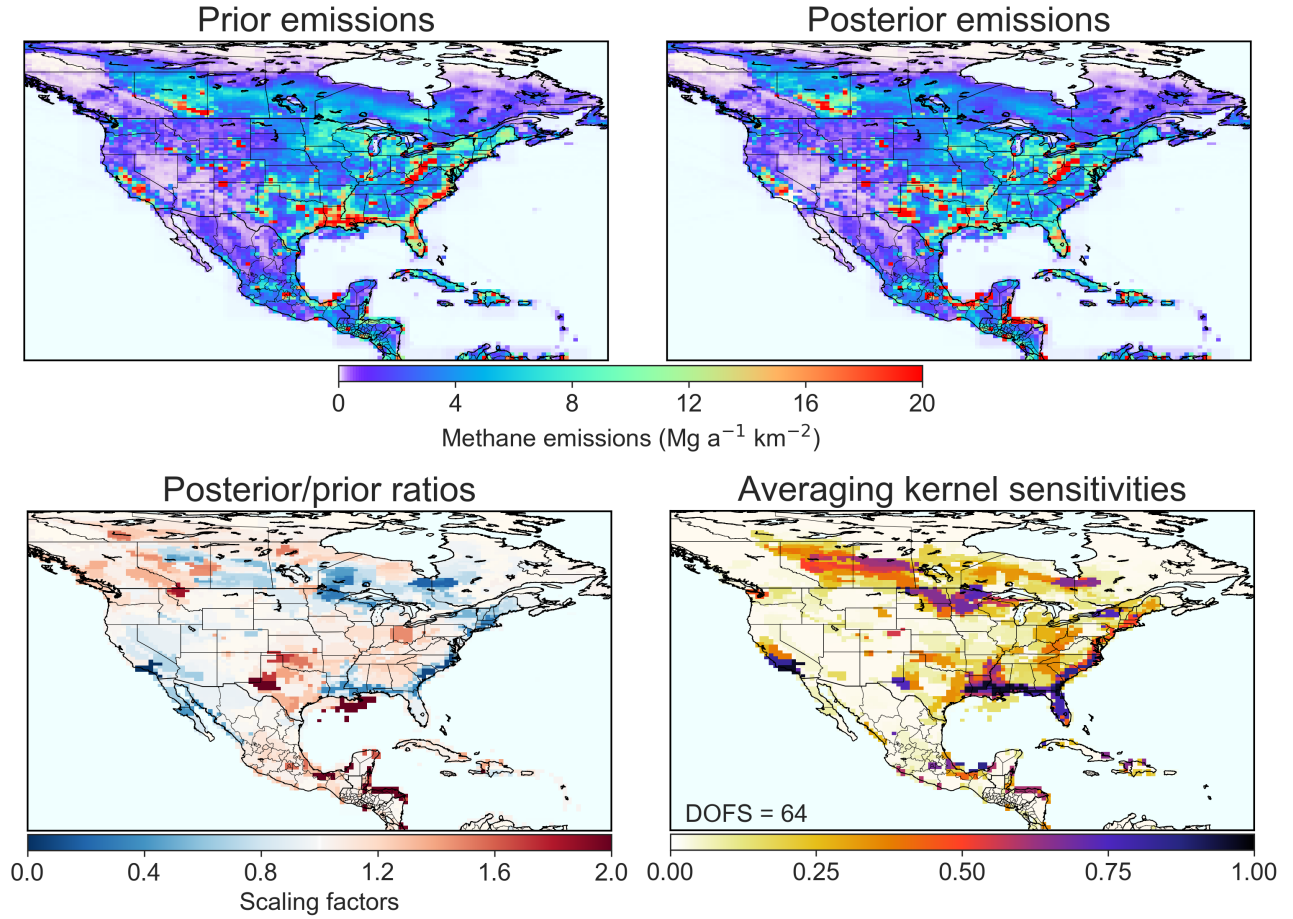


Figure 3. Mean 2010-2015 posterior methane emissions [projected from the 600-member Gaussian mixture to the \$0.5^\circ \times 0.625^\circ\$ model grid](#) and comparison to the prior estimate. Results are from the base inversion. The bottom right panel shows the averaging kernel sensitivities [projected to the model grid](#) (diagonal elements of the averaging kernel matrix). The trace of the averaging kernel matrix, i.e., the degrees of freedom for signal (DOFS), is given inset. It represents the number of independent pieces of information that can be constrained by the inversion.

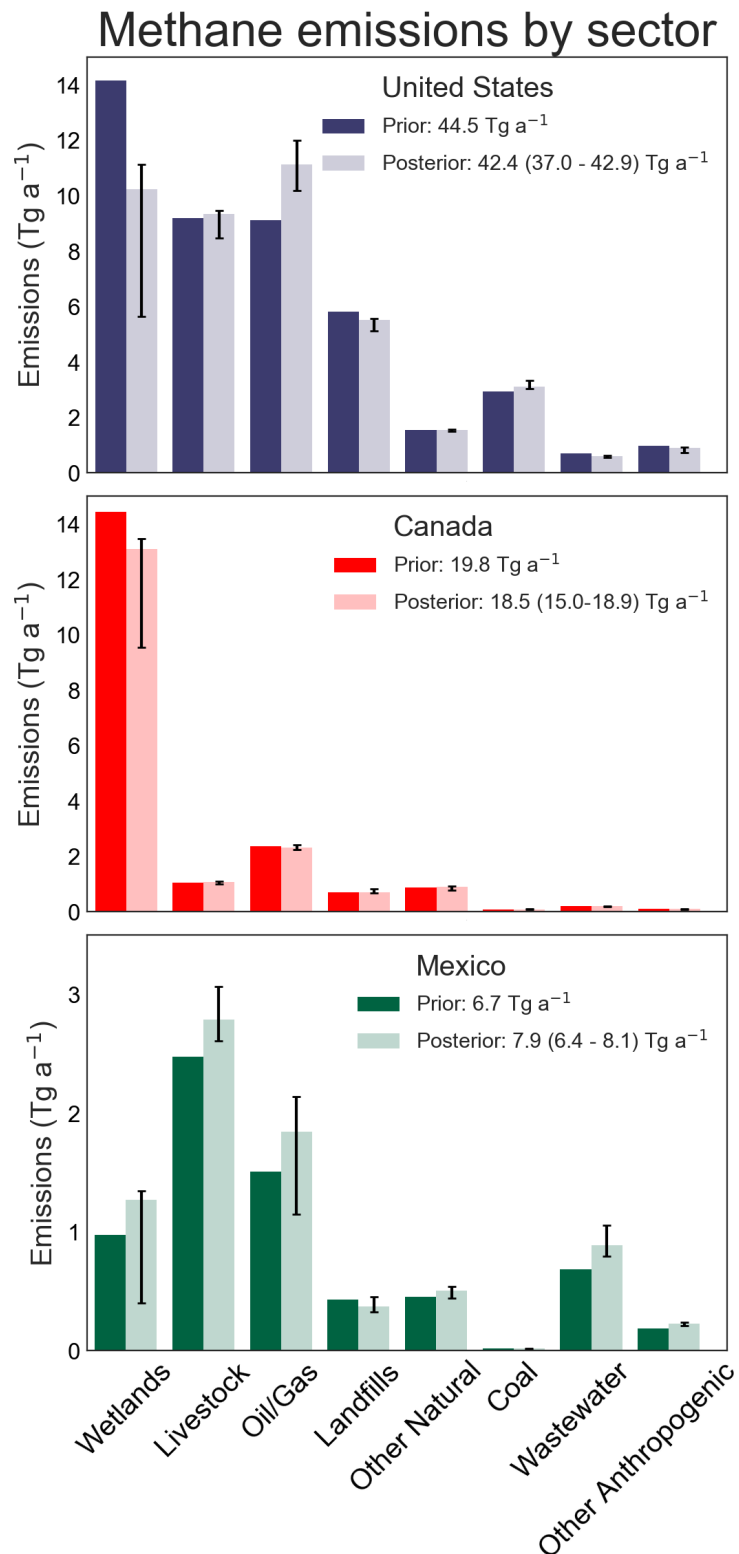


Figure 4. Mean 2010-2015 methane emissions per source sector for the contiguous US (CONUS), Canada, and Mexico. Values are shown for the prior estimates (Table 1) and for the posterior estimates from the after inversion of GOSAT data. Vertical bars show the ranges of results from the inversion ensemble.

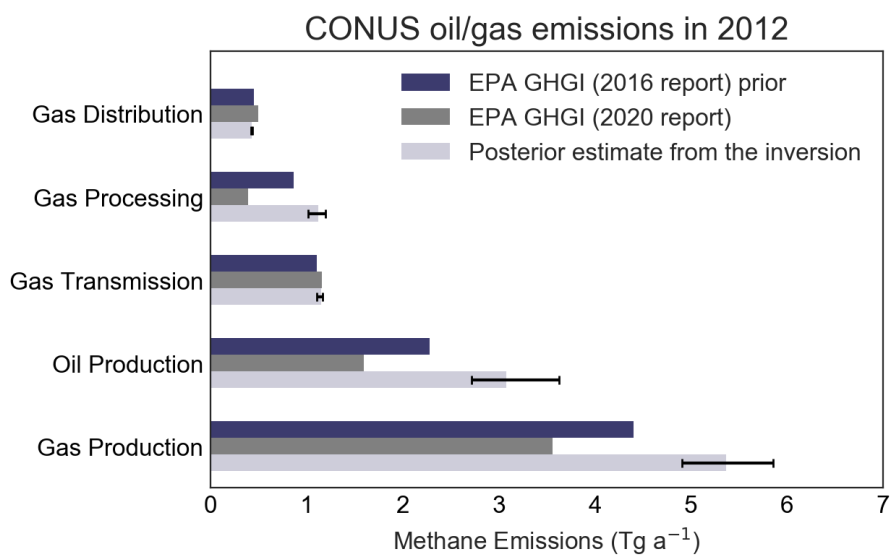


Figure 5. Methane emissions from the oil/gas sector in the contiguous US (CONUS) in 2012. The figure shows the original EPA GHGI estimates for 2012 used as prior in the inversion (EPA, 2016), the updated [EPA GHGI](#) estimates for 2012 based on revised methodology (EPA, 2020), and the posterior results from the inversion. Horizontal bars give the ranges of the inversion ensemble.

Posterior methane emission trends

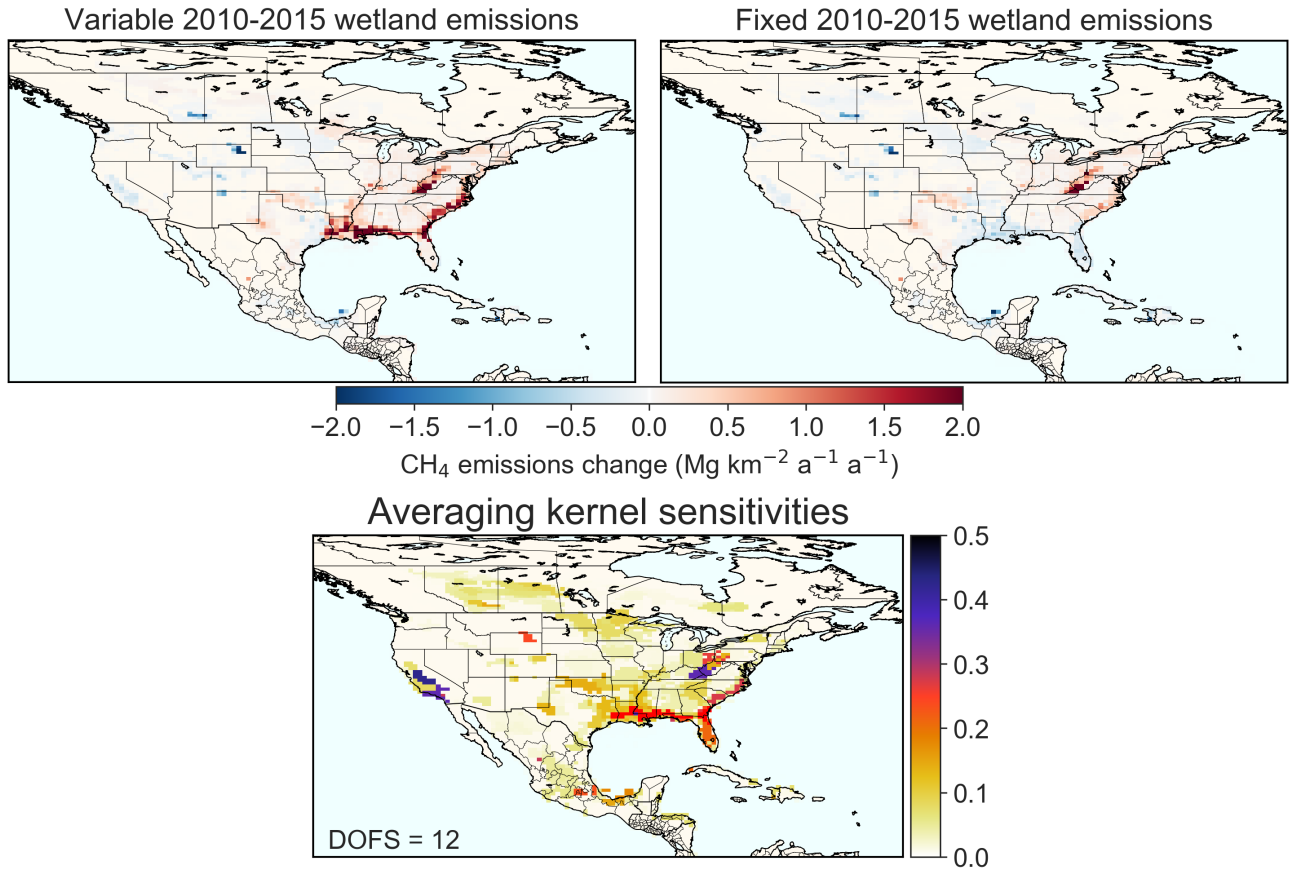


Figure 6. 2010-2015 methane emission trends in North America. The top panels show the posterior estimates from the inversion of GOSAT satellite data, allowing for 2010-2015 interannual variability in the prior estimates for wetlands and open fires (base inversion, left) and ~~assuming constant~~ not allowing for that prior ~~estimates~~ interannual variability (right). The results not allowing for 2010-2015 prior interannual variability of wetland and open fire emissions (right) are more reliable. The bottom figure shows the averaging kernel sensitivities, with the degrees of freedom for signal (DOFS) indicated inset.