We thank the reviewers for their helpful comments that have improved our paper.

Anonymous Referee #1
Received and published: 1 February 2018
<General Comments> Satellite observation is the most important method to provide decade long and global data of anthropogenic methane emission. In 2010s, GOSAT is the only satellite to provide column CH4 density but its spatial coverage is limited and a single data has large fluctuation. Therefore, statistical analysis is important. In addition, selection of reference point together with emission point or estimation of the background is critical for quantitative analysis. This paper proposed and described new analytical method clearly. The trend data from different emission source by this work is innovative. It is worth publication after minor revision.

<Specific Comments>
(1) Page 3, Line 1, Proxy method. "The proxy method uses prior knowledge of carbon dioxide" Brief description of prior knowledge is needed. Does it include seasonal variation plus annual growth only or anomalies such as caused by heat wave in 2010 and El Nino?
We added the description.

(2) Page 3, Line 21 "instrument error" Page 3 line 23 "instrument noise" Page 3 Line 25 "Local instrument bias" Supplemental material, Page 3, Line 12, "instrument error" Supplemental material, Page 4, Figure S2 caption, "instrument noises"
Do these terms have the same meaning? TANSO-FTS onboard GOSAT has two major random error sources and there are also several systematic errors. Detector noise and pointing fluctuation in 4 sec to acquire single interferogram creates random noise. Radiometric calibration error due to degradation after launch, spectral calibration and spectral line shape error, radiative transfer calculation error, molecule parameter cause systematic bias.
Here instrument error/noise means random error, and instrument bias means systematic error. We updated the text accordingly.

(3) Page 5, Line 26, Gulf of Mexico observation by GOSAT "are not directly detectable by GOSAT because the nadir measurements are only over land" It should be described more accurately. Over ocean including Gulf of Mexico, GOSAT can observe column averaged CH4 using glint mode by tracking specular reflection point but the data are sparser.
We added this in the text: "Glint observations are available over the ocean but are much sparser."

<Technical Corrections> (1) Supplemental material, Page 7, Figure S5, Description of blue, black and red lines in the figure caption will help readers' understanding even though they are described in the text.
We added the description in the figure caption.

Anonymous Referee #2
Received and published: 20 March 2018
General comments ———————
This paper is mostly an update of the Turner et al. paper of 2016 aiming at estimating trends in methane emissions over North America as inferred from inversion of GOSAT satellite atmospheric weighted columns. Basically, two more years of data are assimilated and the method to estimate the background is revised. The methodology used here has been criticized in details in Bruhwiler et al (JGR, 2017), main arguments being a too short time window for data assimilation making the GOSAT trends sensitive for instance to changes in atmospheric transport, seasonal biases in GOSAT data towards summer months (less clouds = more data), and influence of the choice of the background. In this paper, the authors address only partly these criticisms and add an original sectorial analysis of the inferred trend.

My main concern on this paper is that it does not fully address the extensive criticisms made in Bruhwiler et al. A window of 6 years is still very short to make a robust trend analysis for a species like methane with a 9-year lifetime and I am not sure that adding 23 months compared to Turner is enough. The inferred trend is very noisy (0.2 ± 0.7 ppb a-1) and moving to percentages is a bit misleading considering the very low value inferred especially when considering the remaining bias of GOSAT data of 4-6 ppb (PVIR4 report from Buchwitz et al., 2016). Nothing seems to be done for the seasonal bias and only the question of backgrounds is addressed in detail. The authors may consider looking at the Cressot et al paper (ACP 2016) on the detectability of emissions at regional scale to figure that trends are very hard to detect with the not-so-dense and biased GOSAT data. The text also lack precision in many places (see specific comments).
Some part of the work in interesting such as the methodology for the sectorial analysis but I think that more time is needed to extent the timeseries and be able to use this approach more safely and provide a reliable update of the Tuner et al. paper addressing all the issues raised since they published it.

Reviewer #2 picks up on the criticisms made by Bruhwiler et al. (2017) of the Turner et al. (2016) paper. Our work has made an honest attempt to address these criticisms (definition of the background, length of the record, inconsistency with surface network) and we have made a good-faith effort to further address the reviewer's concerns in revision. Point-by-point responses are below. It is very doubtful that we can fully satisfy the reviewer but we hope that he/she will let us "agree to disagree" in an open spirit and carry out the discussion in the literature. In answer to the criticisms above:

- **To dismiss the paper as simply an update to Turner et al with two more years of data and different definition of background seems very unfair. This paper adds (1) sectoral breakdown, (2) Canada and Mexico, (3) validation with TCCON, (4) relations of trends to activity data, (5) examination of consistency in the trend with surface sites. These are important advances. In addition, we have extended the GOSAT trend analysis till**

year 2016, by adding an additional year of analysis to what was submitted in the first version to ACPD.

- **Not clear why lifetime is relevant here. The 9-year lifetime of methane is not relevant to the length of the record needed to diagnose a trend. The relevant time scale for a trend in enhancement over background is how long it takes for the enhancement signal to dilute into the background - and that time scale is a few weeks.**

- **The bias in GOSAT data is not relevant since there is no reason to think that it would affect the local background and the enhancement differently (we now make that point in the revised manuscript).**

- **Seasonal bias in the GOSAT data only affects Canada as stated in the text. This doesn't invalidate the trend analysis; it just means that (for Canada) the trend is more of a summertime one.**

We expanded our discussion to address criticisms made in Bruhwiler et al. (see response below and in Specific comments).

Regarding the length of the GOSAT record, we now expand our analysis to 2016 (latest available GOSAT data) for what is now a 7-year record. The addition of 2016 supports the trend previously observed for 2010-2015. We think that a window of 7 years is reasonable to infer methane trends. Lifetime has little to do with it. Methane trend analysis has previously been done using SCIAMACHY with 7 years of data, e.g., Frankenberg et al. (2011). We agree a longer time period would lead to more robust results. We have mentioned this limitation in the conclusion.

Frankenberg, C., I. Aben, P. Bergamaschi, E. J. Dlugokencky, R. van Hees, S. Houweling, P. van der Meer, R. Snel, and P. Tol (2011), Global column-averaged methane mixing ratios from 2003 to 2009 as derived from SCIAMACHY: Trends and variability, J. Geophys. Res., 116, D04302, doi:10.1029/2010JD014849.

Regarding the inferred trend (0.25 ± 0.48 ppb a-1 with the addition of 2016 data). This trend is significant but it is indeed noisy, which is precisely why we move our analysis to the aggregated enhancement. Our conclusion is based on the trends in the aggregated enhancement. We edited our text accordingly (see the response in Specific comment).

The bias in GOSAT data is removed in our approach. We define our local background as low percentiles, and the resulting local enhancement is unbiased as we stated in the text: "This approach removes any local instrument bias (systematic error) because the bias is expected to similarly affect all percentiles of the methane observations."

Regarding GOSAT seasonal bias, we already mentioned this in the text: "GOSAT observes in all seasons with near-uniform frequency south of 45˚N (CONUS and Mexico), but observations further north (Canada) are biased toward summer. The number of successful retrievals over

Canada is 2-3 times less in winter than in summer (see Supplemental Material)." We now mention this explicitly again in the conclusion "… variations in wetland areal extent, though this trend is weighted toward summer because of the seasonal bias in observation frequency (less observations in winter)".

Cressot et al. (2016) found GOSAT performed better than surface observations and IASI for detecting methane anomalies at global and regional scales. The poor rate to detect the methane anomalies at the regional scale as stated by Cressot et al. may be due to that (1) they were conservative to estimate the noise (possibly leading to its overestimation); (2) the time period of GOSAT is 2009-2011, a time period with relatively flat methane signal as seen in our trend analysis. We now mention Cressot et al. in the text.

We do not agree that our text lacks precision in many places (there are 5 places in specific comments related to precision, which are all minor).

Specific comments ————————-

P2 - L10: you may also mention decreasing BBG and quote Worden et al (2018) paper in Nature Comm.
We have added this in the text.

P2 – l14: please add that, contrary to surface networks, the GOSAT data have residual biases of 4-6 ppb as stated in the PVIR reports (Buchwitz et al). Also, the spatial coverage is enhanced by GOSAT but the number of clear-sky scenes is to so huge, and temporal coverage is probably smaller than continuous surface in-situ measurements
We now mention the bias in Methods:
"The resulting GOSAT XCH4 data have been validated against the ground-based Total Carbon Column Observing Network (TCCON), and found to be of high quality with a single-scene precision of 0.7% (random error) and a systematic error of 4-6 ppb (Parker et al., 2015; Buchwitz et al., 2015, 2016)."

We have deleted the text about spatial coverage being enhanced by GOSAT.

P2 – l16-17: there are other reason in Bruhwiler's paper to be added here: impact variations of atmospheric transport linked to short-term window of assimilated data (6- 7 years is still short to me), seasonal bias of GOSAT data. You cannot only pickup what arrange you and have to address all limitations raised by previous work.
Here we updated the text as
"…been biased by the brevity of the GOSAT record, atmospheric transport variability, seasonal bias in GOSAT sampling frequency, and the use of Pacific data as background."

We actually addressed all the limitations later in the text. We now expand these discussions (also see response below).

P2 – l19 : This is not precise enough. short-term trend may depend on local to regional conditions but longer trend is a global signal and one station is enough to get it.
We updated the text as "…local or regional trend detectability from the surface data may be limited by their sparsity".

P2 – l20: lack of precision. which version of EDGAR ? 4.2 has too large emission and trend especially in Asia. EDGAR4.3.2 partly corrects this issue. Please be more precise. Also, the dependency to prior assumption may be loose or tight depending on the associated error structure.
Asia is not relevant here, and EDGAR4.3.2 has its own problems, but we deleted that text as non-essential.

P2 - l22-23 : Adding 2 years compared to Turner et al., 2016 does not convince me that the time period will be long enough to overcome the issues raised in Bruhwiler et al (2017). 10 years (~ methane lifetime) would be a minimum to start extracting reliable information on methane trends to my opinion.
Not clear why lifetime is relevant here. If it was we couldn't say anything about trends of CO_2 on decadal scales…

P3 – l6 : 0.7% is 12 ppb. Are you talking of random error or systematic errors ? please be more precise as systematic errors (estimated at 4-6 ppb from PVIR report of Buchwitz et al) ultimately limit the use of GOSAT to estimate emission trends of a few ppb/yr or less.
As we said in the text, it's instrument precision so here we mean random error. We updated the text as "…a single-scene precision of 0.7% (random error) …". Systematic errors (or bias) are irrelevant for methane enhancement in our approach. We have already discussed this in the text (see P3, L24-26): "… This approach removes any local instrument bias because the bias can be expected to similarly affect all percentiles of the methane observations."

P3 – l9-10 : the opposite is clearly shown in Bruhwiler's paper whith surface emission changes appear only weakly sensitive to surface emissions. Please rephrase.
Replaced "given source region" by "strong source region"

P3 – l16 : " the low (10th -25th) percentiles of the deseasonalized GOSAT methane Observations" unclear to me. Which observations? on which area? how is it specific to the 0.5x0.5 location. Please rephrase to be more clear and explain what you do exactly.
We updated the text as

“Here we define local background methane for a given CONUS location (0.5˚x0.5˚ grid cell, typically including a single repeated GOSAT measurement location) and for a given year as the low (10th-25th) percentiles of the deseasonalized GOSAT methane observations in the given 0.5˚x0.5˚ grid cell and year, …”.

P3 – l20 : how did you choose these upper bopund 25th percentile ? did you try other range and how sensitive is this choice on your results ?
We consider values below 25th percentile to be low percentiles. Results are only weakly sensitive to the choice of different ranges as stated in the text. We also did a sensitivity test on this (see Fig. S5 in supplementary material).

P4 – l3-4 : the trend on enhancements does not seem to be significant considering the error bars. Please provide more quantitative results on this.
Here significance for a single site is not relevant because we only focused on the aggregated enhancement trends. We updated the text as “… although the error standard deviations defined by the ranges of the 10th-25th percentiles are large and the trends at this single site are significant ($p = 0.07$). Below we will use enhancement statistics aggregated over a large number of sites in order to reduce that uncertainty and quantify trends.”

P4-l8: Is EDGAR 4.3.2 very different than 4.2 over North and central America ?
No. They are similar. We added “Compared to EDGAR v4.2, the more recent EDGAR v4.3.2 (Janssens-Maenhout et al., 2017) has similar national totals and spatial patterns for non-oil/gas anthropogenic methane emissions.”

P4 – l19-20: did you try not doing so as it reduces largely the number of wetland- dominated pixels.
Using either wetland inventory alone would bias our results because they differ significantly in space (see Supplement Material). We updated the text as
“Wetland-dominated areas determined by the WETCHIMP mean and WetCHARTs inventories differ significantly (see Supplemental Material). Using either of the two inventories alone may bias our results, and thus we conservatively require wetland-dominated areas to be determined as such in both inventories.”

P4 – l24-25 : what about atmospheric transport ? summing only columns above the high emitting pixels does not account for transport and the potential plume sampling by other GOSAT data. It would be worth mentioning this to clarify what is it you do here.
As we mentioned earlier, the local background range (10th-25th percentiles) accounts for atmospheric transport. We updated the text as:
“To account for background variation due to atmospheric transport, the summation in Equation (1) is conducted for 1000 Monte Carlo realizations where the background $XCH4_{,b,i}$ for each grid cell and for individual years is obtained by random sampling of percentiles in the 10th-25th range.”

P5 – l15 : are they all supposed independant ? How robust is this significance ? Although tighter than in Tiuner et al., the PDF is still broad with a sigma of 0.66

With addition of 2016 the sigma has decreased to 0.48. Each local enhancement is calculated independently. Significance is indicated by p-value <0.01. We agree the sigma is broad, but that's why we move our analysis to the aggregated enhancement that significantly reduced the uncertainty. We now mention that in the text:
"Below we will use the aggregated enhancement (Equation 1) to infer the trends and reduce the uncertainty."

P5 – l16-17 : 10.8 ppb enhancement might be due to other causes as stated in Bruhwiler et al. Please mention that this is a maximum and which of the causes raised in Bruhwiler's paper may still apply here. I strongly recommend to add in the following that inferred numbers are maximum number, potentially smaller because of limitations raised in Bruhwiler's paper.
We do not think that 10.8 ppb is a maximum. As we discussed earlier, local background is statistically not affected by random error, and should account for transport variability to some extent. Janardanan et al. (2017) found a large number of observed and simulated enhancements in the range of 10 to 20 ppb in North America using GOSAT observations and a Lagrangian particle dispersion model. We updated the text accordingly. "The mean 2010 methane enhancement for high-emitting grid cells in CONUS relative to local background is 10.8 ppb, comparable to that found by Janardanan et al. (2017)."

Figure 3 : just ot be sure : the grey bars for wetwhimp and Bloom do reflect the totals for the common pixels ? if not please correct.
No, those are national totals as stated in the figure title and caption, but we now add the totals for common pixels.

P5 – l29 : what about pixerls emitting a lot but with a balanced share of emissions (ivestock & oil&gas) ? Yopur method should discard them. How does it influence your results ?
Here for national trends we do not discard those pixels. We only discard them when we do sectoral trend analysis. So it does not influence national trends. For sectoral analysis, it does not make sense to use grid cells that are not dominated by any source sector. We updated the text as
"Here the trends are calculated for the summed enhancement Δ in Equation (1) calculated for individual years and for high-emitting grid cells in individual countries or high-emitting sectors."

P6 – l1 : replace ambiguous "interannual" by "year-to-year" or equivalent
We replaced it by "year-to-year".

P6 – l14-15 : US oil&gas activity (figure 5) show stalled variations in 2014-15 whereas your analysis find a fast increase from 10 to 20% (figure 4). Isn't that contradictory ? Please comment in the main text.
No, that's not contradictory. In the text, we have already mentioned that "…, though production rate is not necessarily a predictor of emissions (Peischl et al., 2015)."

P6 – l20-22 : how do emission factors for swine and cattle compare ? it would be worth to add the cattle number in comparison with the swine emission factor range given. Is this increase really significant for methane emissions (uncertain range of small emissions of 0.01-0.2 Tg/yr)?
We don't think it's worth to compare cattle population with swine emission factors as it will not convey any new information. We already provided emissions in Midwest from enteric fermentation (cattle) and manure management (swine): "These grid cells emit 0.95 Tg CH_4 a^{-1} from enteric fermentation (cattle) and 0.55 Tg CH_4 a^{-1} from manure management (swine) according to the gridded EPA inventory (Maasakkers et al., 2016)."

This increase is significant and not small for Midwest as we stated in the text that the trend largely reflects Midwest. The uncertainty range here is due to the choice of emission factors.
We added "A larger value of the emission factors is more likely. The emission may increase …"

P6 – l28 : interannual → year-to-year or equivalent
We replaced "interannual" by "year-to-year".

P6 – l30 : " wetland areal extent" : this is very controversial ads there is no consensus of wetland extent and their evolution (see Poulter et al., 2017 also). Please mention this controversy here.
We updated the text as
"…, though the definition of wetland areal extent may vary significantly (Poulter et al., 2017). Here the WetCHARTs extended ensemble used GLOBCOVER land cover data (Bontemps et al., 2011) and the Global Lakes and Wetlands Database (GLWD Lehner and Dölla, 2004) to represent spatial wetland extent, and ERA-interim precipitation to account for temporal wetland extent (Bloom et al., 2017)."

P6 – l33 : please note in the text that the "trend" you infer for CONUS is mostly after 2012 ("total" line on figure 4). The inversions reported in Bruhwiler 2017 stop in 2012. Please mention these two elements in the main text. Again, waiting more time to get longer time series would avoid limitations in trend analysis. . .
We removed Bruhwiler et al. (2017) here. It's irrelevant for our residual test. We updated the text accordingly.

P7 – l1 : Are the stations shown on figure 7 used in the CT inversion? please precise. Do some other surface stations not shown here show some trend? If not please mention it at it reinforce your point.
Yes, they are used in the CTL inversion. We updated the text accordingly.

P7 – l8-9 : But this does not discard the possibility that the trend found in your paper is not due to emissions but to other factors as stated in bruhwiler's paper. Please mention this here as well.
The detected trends have already accounted for the factors as stated in Bruhwiler's paper.

P7-l12 : I recommend to change " significant increase in US methane emissions" into "significant increase in total US methane emissions after 2012"
We changed the text accordingly.

Conclusion : please develop more the main limitations of your study either at the end of result section or in the conclusion.
We added limitations of our study in the text accordingly.

What about OH changes in your method ? you do not mention your assumptions on OH. Please specify them somewhere in the text.
OH is irrelevant here as we already discussed in the text (P3, L28-30): "Any trends in OH concentrations would also not affect the enhancement because the lifetime of methane against oxidation is 9-10 years (Prather et al., 2012; Kirschke et al., 2013), very long compared to the timescale for ventilation from the source region."

~~2010-2015~~ 2010-2016 methane trends over Canada, the United States, and Mexico observed by the GOSAT satellite: contributions from different source sectors

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Abstract. We use ~~six years (2010-2015~~seven years (2010-2016) of methane column observations from the Greenhouse Gases Observing Satellite (GOSAT) to examine trends in atmospheric methane concentrations over North America and infer trends in emissions. Local methane enhancements above background are diagnosed in the GOSAT data on a $0.5° \times 0.5°$ grid by estimating the local background as the low (10^{th}-25^{th}) percentiles of the deseasonalized frequency distributions of the data for individual years. Trends in methane enhancements on the $0.5° \times 0.5°$ grid are then aggregated nationally and for individual source sectors, using information from state-of-science bottom-up inventories~~, to increase statistical power. Our results suggest~~. We find that US methane emissions increased by ~~$2.1 \pm 1.4\%$~~ $2.5 \pm 1.4\%$ a^{-1} (mean $\pm$ one standard deviation) over the ~~six-year~~ seven-year period, with contributions from both oil/gas systems (possibly unconventional oil/gas production) and from livestock in the Midwest (possibly swine manure management). Mexican emissions show a decrease that can be attributed to a decreasing cattle population. Canadian emissions show ~~interannual~~ year-to-year variability driven by wetlands emissions and correlated with wetland areal extent. The US emission trends inferred from the GOSAT data account for about 20% of the observed increase in global methane over the ~~2010-2014 periodbut may be too small to be detectable with surface observations from the North American Carbon Program (NACP) network~~2010-2016 period.

1 Introduction

Methane is an important greenhouse gas with a calculated climate impact as important as carbon dioxide over a 10-year time horizon (Myhre et al., 2013; Etminan et al., 2016). Livestock, oil/gas, and waste are the leading anthropogenic sources. Wetlands are the dominant natural source. Contributions from different source sectors and regions remain poorly quantified (Kirschke et al., 2013; Saunois et al., 2016). Atmospheric methane concentrations leveled off in the 1990s but have been increasing again since 2007 (Dlugokencky et al., 2009). Interpretations of atmospheric observations from surface networks have reached conflicting conclusions as to the cause of the renewed increase, with attributions to (1) natural gas production based on correlation with ethane (Franco et al., 2016; Hausmann et al., 2016; Helmig et al., 2016), (2) agriculture/wetlands based on isotopic information (Nisbet et al., 2016; Schaefer et al., 2016), ~~and~~ (3) reduced biomass burning to reconcile the ethane and isotopic constraints (Worden et al., 2017), and (4) declining concentrations of the OH radical ~~which is~~ (the main methane sink) based on the methylchloroform proxy (Rigby et al., 2017; Turner et al., 2017).

Satellite-based observations of atmospheric methane columns have been available from the TANSO-FTS instrument aboard the Greenhouse Gases Observing Satellite (GOSAT) continuously since May 2009 (Kuze et al., 2016). ~~These satellite data, although still relatively sparse, increase considerably the spatial coverage of methane observations compared to the surface network~~Cressot et al. (2016) found that the GOSAT data had limited success in detecting regional year-to-year trends for 2009-2011. Turner et al. (2016) used GOSAT data from January 2010 to January 2014 to infer a 2.8% a^{-1} increase in methane emissions from the contiguous United States (CONUS), based on the trend in the CONUS enhancement of methane relative to the Pacific Ocean taken as background. Bruhwiler et al. (2017) ~~showed~~ argued that this trend inference could have been biased by the brevity of the GOSAT record~~and by~~, atmospheric transport variability, seasonal bias in GOSAT sampling frequency, and the use of Pacific data as background. They pointed out that global inversions of the surface network data for ~~2000-2014~~ 2000-2012 from the North American Carbon Program (NACP) reveal no significant CONUS emission trend. ~~However, trend detectability from the surface data may be limited by their sparsity. In addition, the inversions rely on prior knowledge of US source patterns from the EDGAR inventory (European Commission, 2011), which is known to have large errors (Maasakkers et al., 2016).~~

Here we reexamine the trend in CONUS emissions implied by the GOSAT data by using a longer record (January 2010 - December ~~2015~~2016), an improved definition of the background that accounts for atmospheric transport variability, and sectoral source information from a new gridded version of the US Environmental Protection Agency (EPA) Greenhouse Gas Inventory (Maasakkers et al., 2016). We ~~evaluate the trends for~~ relate the inferred trends to trends in the underlying activities, and evaluate consistency with trends in the surface network data. We also extend the trend analysis to Canada and Mexico.

2 Methods

GOSAT was launched in January 2009 in a Sun-synchronous low Earth orbit~~, and after 7 years in space it still provides consistent retrieval accuracy (Kuze et al., 2016) of column-integrated methane concentrations. It detects the~~. It retrieves the atmospheric methane column by nadir measurements of solar back-scatter (1.65 μm absorption band). There has been no degradation of retrieval accuracy since the beginning of the record (Kuze et al., 2016). Observations in the standard mode are made at three circular pixels of 10 km diameter across the orbit track 260 km apart, separated by 260 km along the track. The same locations are sampled every 3 days, making for a temporally dense data set at those locations. The observations often switch from the standard mode to focus on targets and this affects the regularity of the sampling.

Here we use the version 7.0 proxy nadir retrievals of GOSAT methane data from Parker et al. (2011, 2015). The proxy method uses prior knowledge of carbon dioxide columns (based on the MACC-II inversion product (v13r2; Chevallier F. et al., 2010) accounting for seasonal and interannual variations) to infer methane column average dry mole fractions X_{CH4} (in ppb) from the ratio of retrieved methane and carbon dioxide columns. ~~This~~ The proxy method takes advantage of the much larger variability in methane than in carbon dioxide mixing ratios (Frankenberg et al., 2006; Parker et al., 2015). The resulting GOSAT X_{CH4} data have been validated against the ground-based Total Carbon Column Observing Network (TCCON), and found to be of high quality with a single-scene precision of 0.7% ~~(Buchwitz et al., 2015; Parker et al., 2015)~~(random error) and a systematic error of 4-6 ppb (Parker et al., 2015; Buchwitz et al., 2015, 2016). GOSAT observes in all seasons with near-uniform frequency south of 45°N (CONUS and Mexico), but observations further north (Canada) are biased toward summer. The number of successful retrievals over Canada is 2-3 times less in winter than in summer (see Supplemental Material).

From a simple mass balance perspective, enhancements of column methane above the surrounding background in a ~~given~~ strong source region can be linearly related to the emissions in that region (Jacob et al., 2016; Buchwitz et al., 2017). Turner et al. (2016) estimated the CONUS background by using glint mode retrievals from GOSAT over the Pacific Ocean for the corresponding latitudes. Bruhwiler et al. (2017) pointed out that changes in large-scale meridional transport could alias trends in this background estimate onto trends in the emissions.

Here we define local background methane for a given CONUS location ($0.5° \times 0.5°$ grid cell, typically including a single repeated GOSAT measurement location) and for a given year as the low (10^{th}-25^{th}) percentiles of the deseasonalized GOSAT methane observations within the given $0.5° \times 0.5°$ grid cell, with seasonality removed using the seasonal-trend loess (STL) decomposition method (Cleveland et al., 1990). This approach assumes that the low percentiles of concentrations reflect meteorological conditions where local sources have relatively little effect on methane concentrations due to rapid ventilation. ~~It allows definition of local enhancements relative to a regional background and this will be important for our sectoral attribution that follows.~~ Low percentiles are a standard approach for estimating the regional background at a measurement location (Goldstein et al., 1995). By choosing the 10^{th}-25^{th} percentile rather than a lower extreme we guard against the effect of ~~instrument error or anomalous flow conditions (such as incursions of tropical air~~measurement noise (random error). A permutation resampling test shows that GOSAT observations across North America are sufficiently precise that ≥10th percentiles are not affected by ~~instrument~~ measurement noise (see Supplemental Material). We use the range defined by the 10^{th}-25^{th} percentile range as a measure of uncertainty in the background for purpose of determining the enhancement. ~~This approach also removes any local instrument bias~~

Systematic errors of 4-6 ppb in GOSAT observations (Buchwitz et al., 2016) do not affect the enhancement because the bias can be expected to similarly affect all percentiles of the methane observations. Local enhancements are inversely proportional to wind speed (Jacob et al., 2016), but we find no significant trends in wind speeds over the ~~2010-2015~~ 2010-2016 period that would contribute to our aggregated trends in methane enhancements (see Supplemental Material). Any trends in OH concentrations would also not affect the enhancement because the lifetime of methane against oxidation is 9-10 years (Prather et al., 2012; Kirschke et al., 2013), very long compared to the timescale for ventilation from the source region.

We examined the validity of our approach by comparing frequency distributions of GOSAT methane columns and related trends to continuous ground-based column observations available from the TCCON (Wunch et al., 2011) network site at Lamont, Oklahoma (36.6°N, 97.4°W). Figure 1 shows the frequency distributions of the deseasonalized GOSAT and TCCON observations at Lamont. The GOSAT background defined by the 10^{th}-25^{th} percentiles is consistent with TCCON; we see that the repeated observation strategy of GOSAT at its discrete sampling locations makes for a sufficiently dense data set for defining the 10^{th}-25^{th} percentiles with little effect from instrument noise. The local annual mean background increases between 2010 and 2015 in a consistent way in the GOSAT and TCCON data sets, reflecting the global increase in the methane background. The enhancements above background also show comparable ~~2010-2015~~ 2010-2016 trends between the two data sets, although the error standard deviations defined by the ranges of the 10^{th}-25^{th} percentiles are large ~~. Here~~ and the trends at this single site are marginally significant ($p = 0.07$). Below we will use enhancement statistics aggregated over a large number of sites in order to reduce that ~~error~~ uncertainty and quantify trends.

To aggregate trends in methane enhancements ~~over different~~ for individual source sectors, we use bottom-up annual mean sectoral information with $0.1° \times 0.1°$ spatial resolution from the gridded 2012 US EPA inventory of Maasakkers et al. (2016), the 2013 Canadian and 2010 Mexican oil/gas emission inventories of Sheng et al. (2017), and the EDGAR v4.2 global inventory for 2008 (European Commission, 2011) for other Canadian and Mexican sources. Compared to EDGAR v4.2, the more recent EDGAR v4.3.2 (Janssens-Maenhout et al., 2017) has similar national totals and spatial patterns for non-oil/gas anthropogenic methane emissions in North America. For wetlands, we use multiyear annual mean values from two climatological inventories with $0.5° \times 0.5°$ spatial resolution: (1) the mean of inventories contributing to the Wetland CH_4 Inter-Comparison Of Models Project (WETCHIMP) (Melton et al., 2013), and (2) the 2010-2015 mean of the WetCHARTs extended ensemble wetland methane emissions inventory by Bloom et al. (2017). From these inventories we select high-emitting grid cells at $0.5° \times 0.5°$ resolution ~~(equivalently about 55 by 45 km resolution in the central Oklahoma)~~ dominated by a particular source sector. The high-emitting grid cells are defined as having emissions larger than 0.5 tons h^{-1}, encompassing 80-90% of anthropogenic and wetland emissions in all three countries. A high-emitting grid cell is identified as dominated by a given source sector if that source sector accounts for more than 70% of the total emissions in the cell. This allows us to define grid cells dominated specifically by oil/gas, livestock, waste, and wetlands emissions. Contributions from other sectors (up to 30%) may lead to some smoothing of results. Wetland-dominated areas determined by the WETCHIMP mean and WetCHARTs inventories differ significantly (see Supplemental Material)~~, and here~~ . Using either of the two inventories alone may bias our results, and thus we conservatively require wetland-dominated areas to be determined as such in both inventories.

We define a total methane enhancement Δ for a given year, source sector, and country as

$$\Delta = \sum_i \left(\bar{X}_{CH4,i} - X_{CH4,b,i} \right), \tag{1}$$

where $\bar{X}_{CH4,i}$ is the annual mean value of the deseasonalized column average dry mole fractions in $0.5° \times 0.5°$ grid cell i for the given year, $X_{CH4,b,i}$ is the corresponding local background value, and the summation is over all high-emitting grid cells for that sector and country. We require grid cells to have at least eight valid retrievals for a given year, and about 70% of grid cells meet this requirement. ~~The~~ To account for local background variation due to atmospheric transport, the summation in Equation (1) is conducted for 1000 Monte Carlo realizations where the background $X_{CH4,b,i}$ for each grid cell and for individual years is obtained by random sampling of percentiles in the 10th-25th range. Results are only weakly sensitive to the choice of that range (see Supplemental Material). The resulting summation statistics define the probability density function of the total enhancement Δ, and this is used in what follows to test the statistical significance of year-to-year trends in Δ.

3 Results and discussion

Figure 2 (upper panel) shows the spatial distribution of GOSAT methane trends in local enhancements over North America at $0.5° \times 0.5°$ spatial resolution from January 2010 to December ~~2015 (six~~ 2016 (seven years of data). The $0.5° \times 0.5°$ trends are inferred from ordinary least-square linear regression of the enhancements for individual years. The trends are not statistically significant at that resolution. We will aggregate grid cells in what follows to increase statistical significance. Some areas are sparsely sampled, such as California, while the central US is more densely observed due to a more regular schedule of standard measurements. Spatial averaging to $4° \times 4°$ as in Turner et al. (2016) does not improve significance (see Supplemental Material) because methane emissions are not correlated on that scale. A major reason for the weaker statistical significance of our results relative to Turner et al. (2016) is the choice of background. Enhancements defined relative to the Pacific background, as in Turner et al. (2016), ~~integrate emission influences over a broader spatial footprint~~ are larger than in our approach where the background is defined locally.

We improve the statistical significance of the CONUS enhancement trends by taking national statistics over all $0.5° \times 0.5°$ grid cells. This is shown in the lower panels of Figure 2 with the CONUS frequency distribution of trends in mean methane, local background, and the enhancements computed by difference. The mean ~~2010-2015~~ 2010-2016 trend in methane enhancements over CONUS is ~~0.21 ± 0.66~~ 0.25 ± 0.48 ppb a^{-1} (mean $\pm$ one standard deviation), which is statistically significant (sample size ~~$n = 254$~~ $n = 241$ and p-value < 0.01). The mean 2010 methane enhancement for high-emitting grid cells in CONUS relative to local background is 10.8 ppb, comparable to that found by Janardanan et al. (2017). If this mean enhancement is taken as a measure of CONUS emissions, then a ~~0.21~~ 0.25 ± 0.48 ppb a^{-1} trend implies a ~~1.9%~~ $2.3 \pm 4\%$ a^{-1} increase in emissions for ~~2010-2015~~2010-2016. The Turner et al. frequency distributions, shown in the lower left panel, are much broader than ours because they did not use annual averaging of the data. Their Pacific background distribution is similarly broader and is also lower than our local background, which is appropriately elevated by continental influences. Below we will use the aggregated enhancements by source sectors (Equation 1) to infer the trends and reduce the uncertainty.

Figure 3 shows the locations of high-emitting $0.5° \times 0.5°$ grid cells dominated by different sectors as identified by the bottom-up inventories of Section 2. Also shown are national emission totals from these inventories. Wetland-dominated areas in Figure 3 are those identified by both the WETCHIMP mean and Bloom et al. (2017) inventories in order to avoid false positives. There is clear separation of grid cells dominated by wetlands, oil/gas, and livestock source sectors. Waste emissions dominate in urban areas but are more localized. Offshore oil/gas emissions over the Gulf of Mexico account for more than 50% of Mexican oil/gas total (Sheng et al., 2017), but are not directly detectable by GOSAT ~~because the nadir measurements are only~~ nadir measurements over land. Glint observations are available over the ocean but are much sparser.

Figure 4 shows GOSAT methane enhancement trends for ~~2010-2015~~ 2010-2016 (expressed as percent change since 2010) over Canada, CONUS, and Mexico, along with contributions from the sector-resolved high-emitting grid cells. Here the trends are calculated for the summed enhancement Δ in Equation (1) calculated for individual years and for high-emitting grid cells of individual countries or high-emitting sectors. Inferring significant trends for a given source sector generally requires $\sim$50 contributing $0.5° \times 0.5°$ grid cells. The largest source of uncertainty is the selection of the local background within the 10$^{\text{th}}$-25$^{\text{th}}$ percentile range, and this is reflected by the error bars in the figure.

The Canadian methane emissions show no significant ~~six-year~~ seven-year trend but large ~~interannual~~ year-to-year variability driven by wetlands. The 2014 maximum can be explained by a maximum of wetland areal extent (Bloom et al., 2017) (See Fig. S6 in Supplemental Material). Observations in the oil/gas dominated region of Canada (mainly natural gas in Alberta) are too sparse for inferring a significant oil/gas emission trend and are not shown here.

Mexican national emissions (excluding oil/gas offshore emissions) show a 5-10% decrease over the 2010 to ~~2015~~ 2016 period that appears to be largely driven by livestock. The decrease of livestock emissions ($4.0\pm1.6\%$ a^{-1}) is consistent with the 17% decrease in the Mexican cattle population over that period as reported by the Foreign Agriculture Service of the US Department of Agriculture (2015) and shown in Figure 5. The slight increase in Mexican emissions from 2012 on suggests an increasing source to compensate for the declining livestock emissions but GOSAT observations are too sparse to identify that source.

The CONUS data imply a significant increase in methane emissions from 2010 to ~~2015~~2016, with a trend of ~~2.1~~2.5$\pm1.4\%$ a^{-1} derived from linear regression that is consistent with our previously calculated mean trend of ~~1.9~~2.3% a^{-1} averaged over the $0.5° \times 0.5°$ gridded trends in Figure 2. Breakdown by sector suggests that US oil/gas emissions increased at a marginally significant level (~~3.1~~2.9% a^{-1}, ~~$p = 0.08$~~$p = 0.03$) from 2010 to 2015. Oil and unconventional (hydraulic fracturing) gas production grew by 15% a^{-1} and 19% a^{-1}, respectively during that period (Figure 5), though production rate is not necessarily a predictor of emissions (Peischl et al., 2015).

The US livestock emissions show a ~~$3.6 \pm 2.3\%$~~ $3.5 \pm 1.8\%$ a^{-1} increase in our analysis, largely reflecting the agricultural Midwest where high-emitting grid cells are concentrated (Figure 3). These grid cells emit 0.95 Tg CH4 a^{-1} from enteric fermentation (cattle) and 0.55 Tg CH4 a^{-1} from manure management (swine) according to the gridded EPA inventory(Maasakkers et al., 2016). The cattle population in that region does not show a significant trend (Figure 5), but swine population in Iowa (accounting for most of the swine population in the Midwest) increased by two million heads from 2010 to ~~2015~~2016(USDA National Agricultural Statistics Service, 2015b; Iowa Department of Natural Resources, 2017) (Figure 5). This would increase swine manure management emissions by 0.02-0.1 Tg CH4 a^{-1} over the ~~2010-2015~~ 2010-2016 period assuming the IPCC (2006) emission factor of 10-45 kg CH4 head^{-1} a^{-1}. Here a larger value of the emission factor is more likely. The emission factor may ~~also~~ have increased during that time due to an increase in swine body weight and a 30% rise in concentrated animal feeding operations (CAFOs) with more than 1,000 animal units (Iowa Department of Natural Resources, 2017). Those CAFOs tend to use liquid manure storage (US EPA, 2016) and have extended manure storage time (Iowa Department of Natural Resources, 2011), which lead to greater methane emissions. A recent bottom-up study from Wolf et al. (2017) found a steady increasing trend since the 1990s in US methane emissions from manure management.

US wetlands emissions do not show a significant trend over ~~2010-2015 but large interannual~~ 2010-2016 but large year-to-year variability, which contributes in part to the total national trend after 2012. Correlation with driving variables in the WetCHARTs yearly ensemble of Bloom et al. (2017) suggests that this ~~interannual~~ year-to-year variability is related to wetland areal extent, same as for Canada (See Fig. S6 in Supplemental Material)~~.~~, though the definition of wetland areal extent may vary significantly (Poulter et al., 2017). Here the WetCHARTs extended ensemble used GLOBCOVER land cover data (Bontemps et al., 2011) a the Global Lakes and Wetlands Database (GLWD; Lehner and Dölla, 2004) to represent spatial wetland extent, and ERA-interim precipitation to account for temporal wetland extent (Bloom et al., 2017).

Inverse analyses of methane concentrations in surface air measured as part of the North American Carbon Program (NACP; Wofsy and Harris, 2002) for 2010-2014 reveal no significant trends in US emissions over that period ~~(Benmergui et al., 2015; Bruhwiler et a~~ We examined whether the trends inferred from this work (significant trends after 2012) are consistent with the information provided by NACP surface data. For this purpose, we examined the residuals (observed minus simulated methane concentrations) of the CarbonTracker-Lagrange (CT-L) methane transport model (Benmergui et al., 2015) driven with two sets of emissions (1) the CT-L posterior emissions for 2010-2014 that are optimized to match all NACP data and show no significant trend, and (2) a scaled version of the CT-L posterior emissions that matches the sector-resolved trends derived in this work. Figure 7 shows annual statistics and trends of the residuals for both simulations at three NACP sites included in the CT-L inversion: LEF (Park Falls, Wisconsin, 45.9°N, 90.3°W), WBI (West Branch, Iowa, 41.7°N, 91.4°W), and WKT (Moody, Texas, 31.3°N, 97.3°W). These sites are strongly influenced by large livestock/wetlands, livestock~~and,~~ , and oil/gas sources, respectively (Benmergui et al., 2015). There is no significant trend in the residuals of the CT-L simulation driven by either our GOSAT-inferred emission trends or CT-L posterior emissions, and the two sets of residuals are statistically indistinguishable. We find similar results for other NACP sites that are less sensitive to source regions. This implies that the trends found in this work are compatible with the constraints provided by NACP data. This also suggests that the surface data may be spatially too sparse to adequately infer trends of the magnitude ~~as~~ detected by GOSAT.

4 Conclusions

In conclusion, analysis of ~~six years (2010-2015~~seven years (2010-2016) of GOSAT methane trends over Canada, the contiguous US (CONUS), and Mexico suggests a significant increase in total US methane emissions after 2012 and decrease in Mexican emissions. The Mexican decreasing trend appears to be due to a declining cattle population. Canada shows no significant long-term trend but large ~~interannual~~ year-to-year variability associated with wetlands and correlated with variations in wetland areal extent, though this trend is weighted toward summer because of the seasonal bias in observation frequency (less observations in winter). The US trend is ~~$+2.1 \pm 1.4\%$~~ $+2.5 \pm 1.4\%$ a^{-1} for the period and appears to reflect contributions from both oil/gas and livestock. Assuming ~~38-53~~ 38-55 Tg CH4 a^{-1} for the CONUS emissions~~(European Commission, 2011; Melton et al., 2013; Turner et al., 2~~ ~~this implies~~ , including 29-40 Tg CH4 a^{-1} from anthropogenic sources (Miller et al., 2013; Wecht et al., 2014; Turner et al., 2015; Maasakk 9-15 Tg CH4 a^{-1} from wetlands (Melton et al., 2013; Bloom et al., 2017), we deduce an increasing emission trend of ~~0.8-1.1~~ 0.9-1.3 Tg CH4 a^{-1} over the ~~2010-2014~~ 2010-2016 period, which would account for about 20% of the global increase in atmospheric methane (Rigby et al., 2017). Our analysis is mainly limited by the length of GOSAT record, and a longer record can provide more reliable results. The definition of local background may also not fully account for the variation in atmospheric transport. Our trend analysis should be compared to trends inferred from inverse modeling (Bruhwiler et al., 2017), which better account for the role of atmospheric transport but have their own errors notably in the prior assumptions of emission patterns (Maasakkers et al., 2016). ~~Better~~ Future inversions combining GOSAT and surface network data with improved bottom-up ~~understanding of the factors driving methane emissions and the implications for trends is ultimately needed~~ estimates are needed to provide more robust trend analyses.

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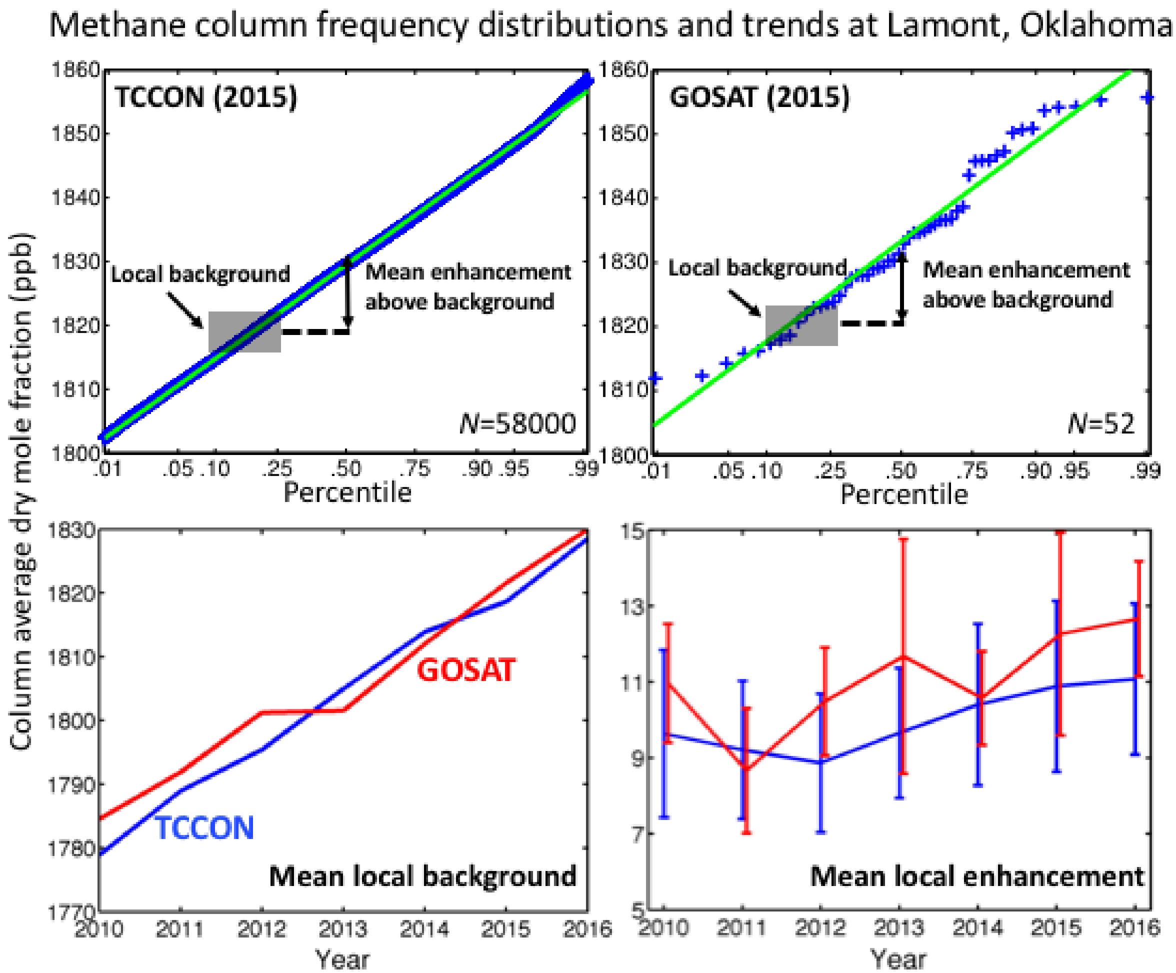

Figure 1. Frequency distributions and ~~2010-2015~~ 2010-2016 trends of methane column average dry mole fractions X_{CH4} at Lamont, Oklahoma (36.6°N, 97.4°W) as measured by TCCON and GOSAT. The upper panels show the deseasonalized 2015 frequency distributions from TCCON and GOSAT. The percentiles are plotted on a normal probability scale such that a normal distribution would plot as a straight line. The local background is defined by the 10^{th}-25^{th} percentile range and the mean annual local enhancement relative to this background is defined by the difference with the mean of the distribution. Lower panels compare TCCON and GOSAT backgrounds and enhancements for ~~2010-2015~~2010-2016, with error standard deviations on the enhancements as described in the text.

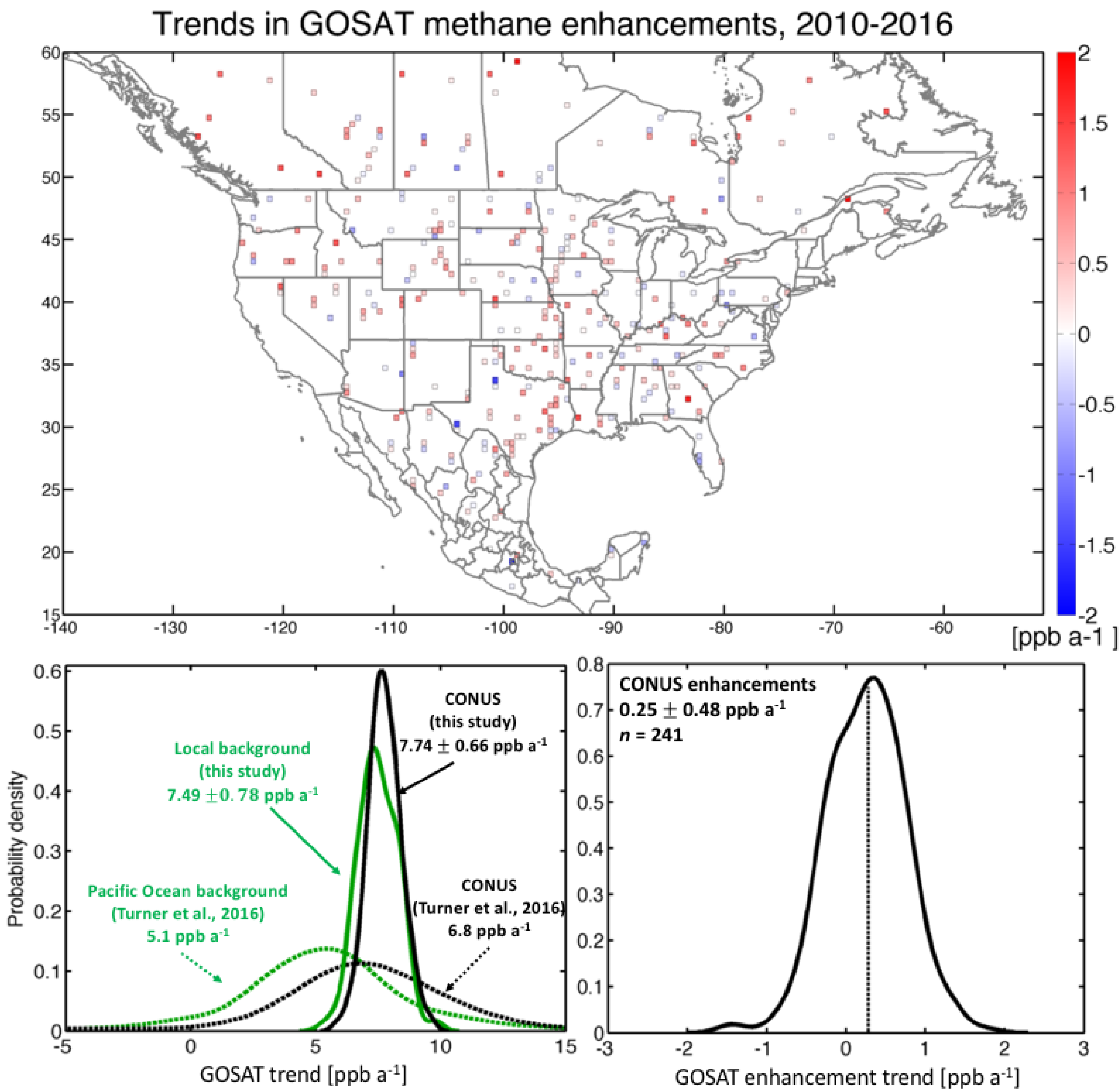

Figure 2. ~~2010-2015~~ 2010-2016 trends in GOSAT methane enhancements over North America. Upper panel: ordinary least-square linear regression trends for $0.5° \times 0.5°$ grid cells with sufficient GOSAT observations, where the deseasonalized annual mean methane enhancements are defined relative to a local low-percentile background as described in the text. The trends are not statistically significant at that resolution (see text). Lower panels: spatial frequency distributions for the $0.5° \times 0.5°$ grid cells over the contiguous United States (CONUS) of mean methane and local background (at left), and local methane enhancements computed by difference (at right). The dashed black line in the lower right panel indicates the mean trend in CONUS enhancements. Also shown in the lower left panel are the 2010-2013 trend distributions from Turner et al. (2016).

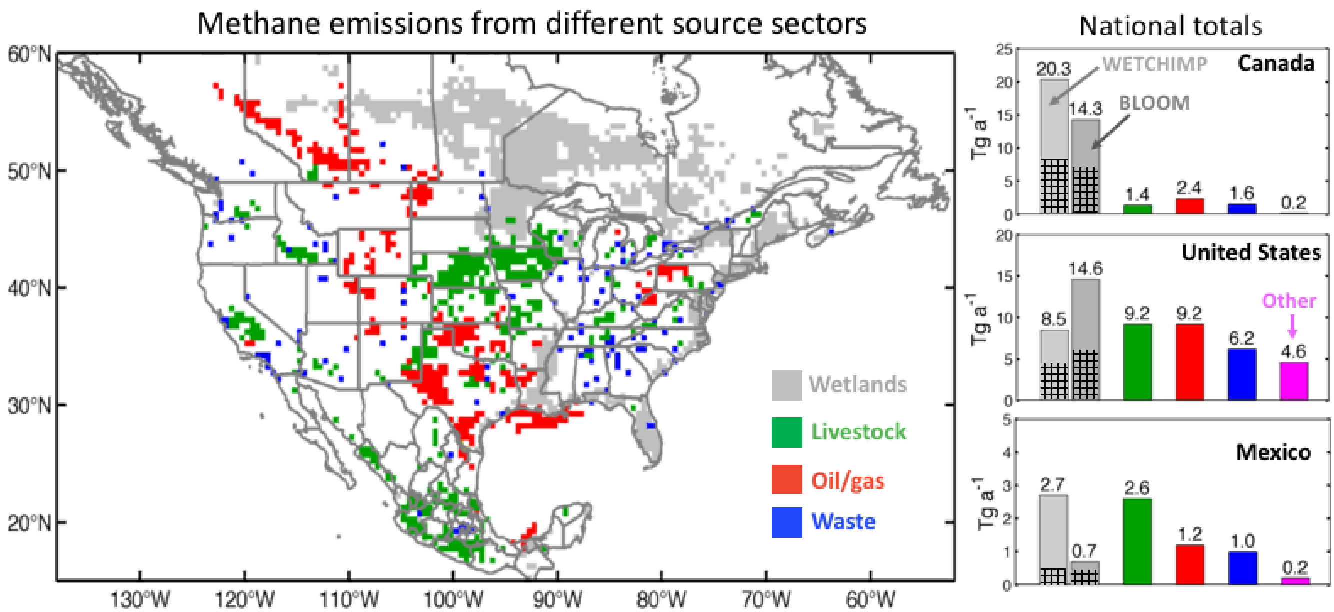

Figure 3. Methane emissions in North America and contributions from different source sectors. The left panel shows $0.5° \times 0.5°$ grid cells with high emissions dominated by a particular sector as identified by the bottom-up inventories (see text for details). High-emitting wetland areas are those identified by both the WETCHIMP mean inventory and the Bloom et al. (2017) mean inventory. Livestock includes enteric fermentation and manure management. Oil/gas includes the complete systems from production to distribution. Waste includes landfills and wastewater plants. The right panel shows national emissions for 2008-2013 from the bottom-up inventories. "Other" includes smaller sources from coal, rice, combustion, petrochemical production, ferroalloy production, and biomass burning. Total emissions in the high-emitting wetland areas are indicated by gridded areas.

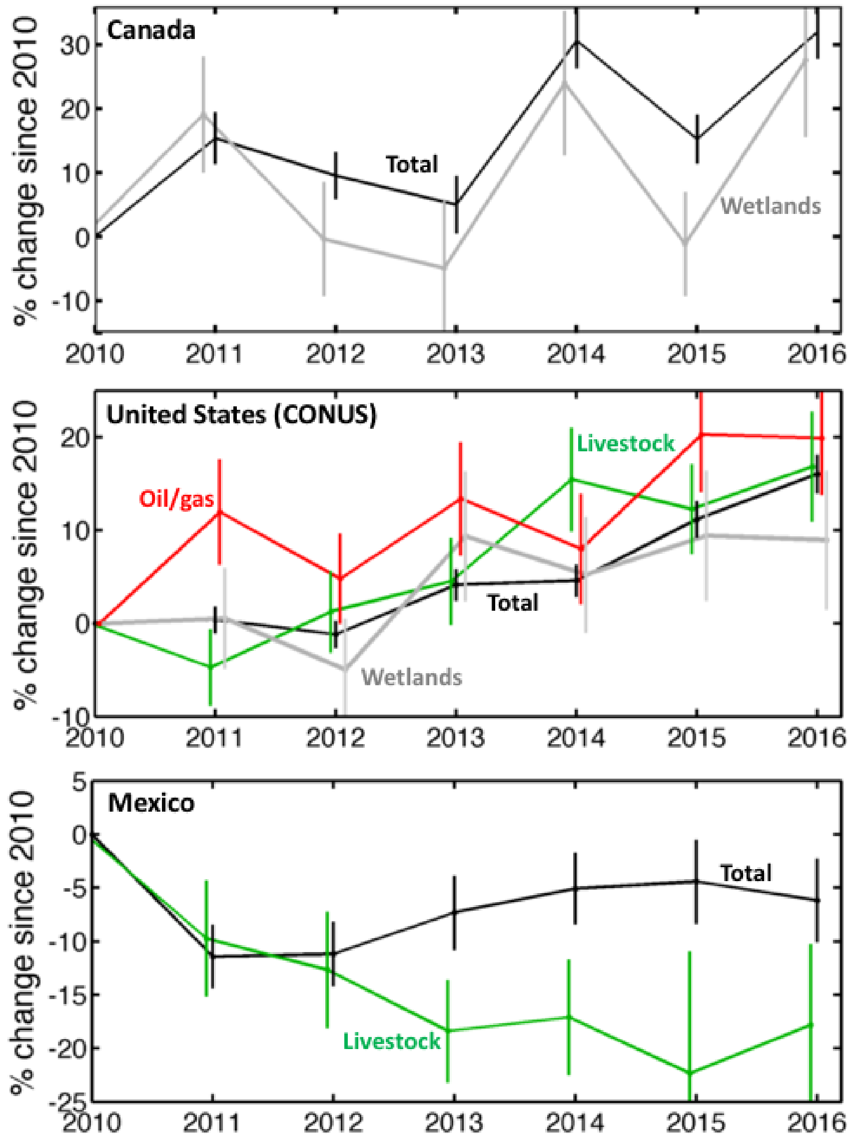

Figure 4. National trends in methane emissions since 2010 inferred from GOSAT, and contributions from specific source sectors where sufficient data are available. The trends are defined by relative year-to-year changes in the summed methane enhancements Δ relative to the local backgrounds as computed from Equation (1), and vertical bars are standard deviations derived from uncertainty in the local background (see text).

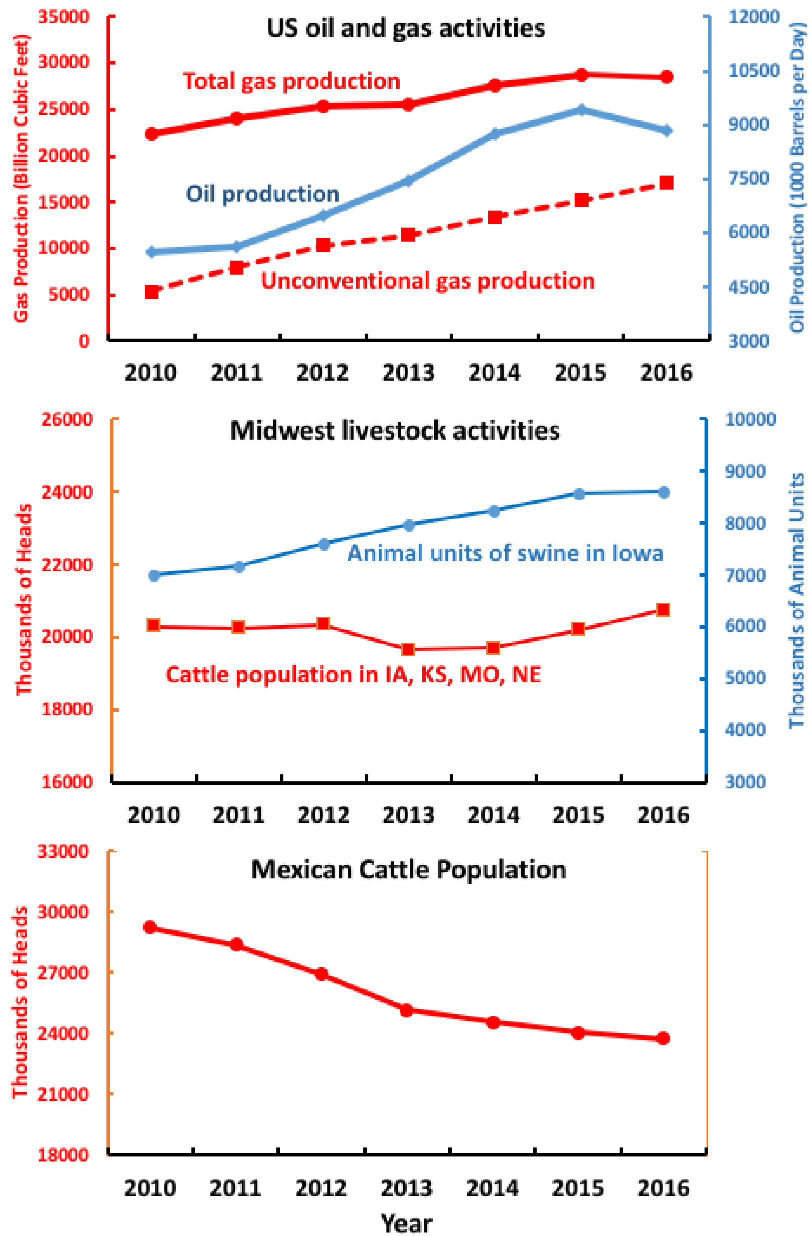

Figure 5. ~~2010-2015~~ 2010-2016 changes in methane emitting activities. Upper panel: ~~monthly~~ oil and natural gas production in CONUS (Drillinginfo, 2016). Middle panel: cattle population in Iowa, Kansas, Missouri, and Nebraska (USDA National Agricultural Statistics Service, 2015a), and animal units of swine in Iowa (Iowa Department of Natural Resources, 2017). One animal unit accounts for 3-5 heads of swine depending on body weight(USDA National Agricultural Statistics Service, 1995). Lower panel: total cattle population in Mexico (USDA Foreign Agricultural Service, 2015).

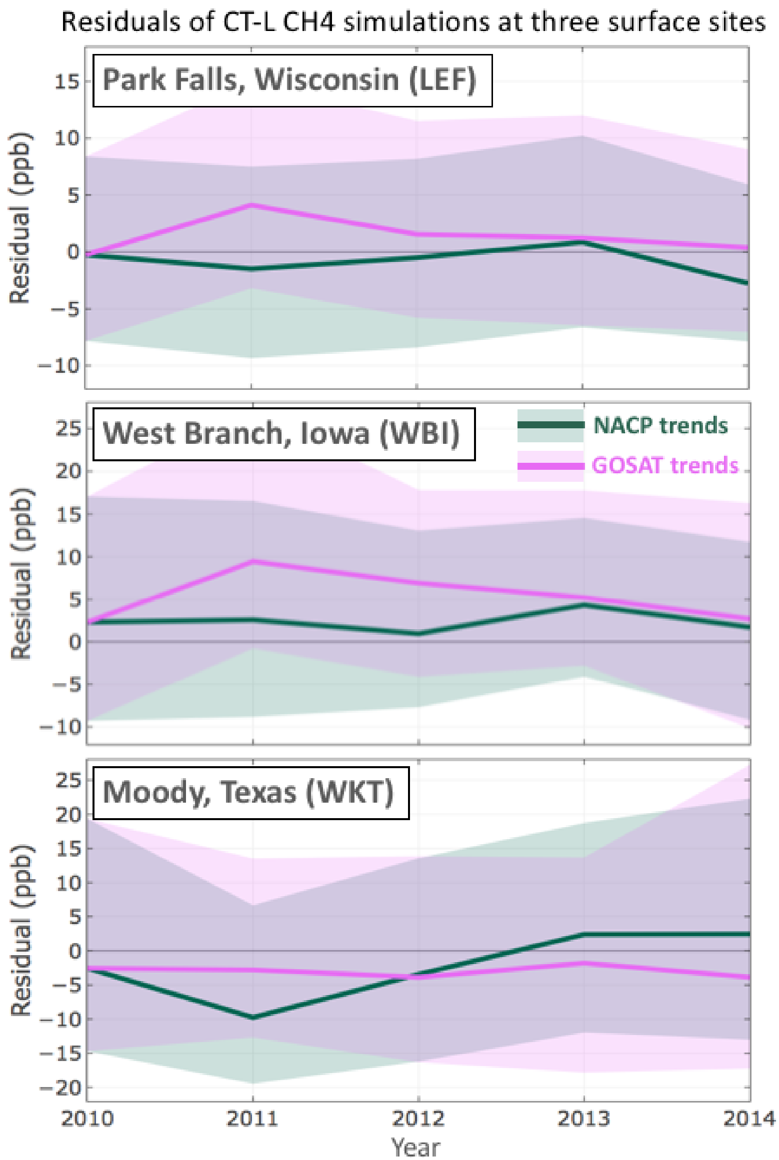

Figure 6. Time series of the residuals (observed minus simulated methane concentrations) of the CarbonTracker-Lagrange (CT-L) CH4 transport model simulations driven by posterior emissions optimized for NACP data (green) and scaled to GOSAT-inferred emission trends (purple) for three surface sites particularly sensitive to emissions from different sectors: LEF (45.9°N, 90.3°W), WBI (41.7°N, 91.4°W), and WKT (31.3°N, 97.3°W). Solid lines show the medians of NACP and GOSAT trends, and shaded areas show the 25th-75th percentile envelope.

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