Reply to Comments by Referee #2

We thank the referee for the time spent on commenting on the paper, and for the encouraging comments and useful suggestions which have helped improve the paper. Below are our responses; the referee's comments are copied *in italic and red*. In the responses, we also indicate the changes made in the manuscript in blue.

The paper deals with the estimation of Canadian methane emissions for the period 2007-2017. It is a valuable contribution to the topic, performing an ensemble regional inversion constrained with the Environment and Climate Change Canada (ECCC) surface measurement network and provides a new perspective on the important region of Canada, where large uncertainties in the methane budget have previously been recognised. The modelling methodology is well established and the ECCC data are widely used and of high quality. A couple of questions remain to be clarified:

1. *l 15: what are the gradients mentioned here?*

By "gradients", we mean "the difference of atmospheric CH4 from one site to another". Line 15 has been changed as follows:

First, the modelled differences in atmospheric CH_4 among the sites show improvement after inversion when compared to observations, implying the CH_4 observation differences could help verify the inversion results.

2. *l 50: could maybe mention that some studies indicate a decreasing methane emission trend in the future due to increased evapotranspiration and drying of the soil (Kwon et al, 2022, https://onlinelibrary.wiley.com/doi/10.1111/gcb.16394)*

Thank you for your suggestion. We included the reference, as below:

Recent studies on the arctic and boreal peatlands reveal more complex sensitivities of CH_4 flux exchange. Kwon et al. (2022) noted a decreasing methane emission trend in the future due to increasing evapotranspiration and drying of the soil.

3. *105: LacLaBiche, Egbert and Downsview still have high variability after data selection. I wonder how you ensure that this is not very local influence, as, if I am not wrong, Egbert and Downsview appear to be within an urban area. Did you use any additional selection methods for these sites, e.g. wind speed?*

We did not use any additional selection to these three sites, Lac La Biche, Egbert and Downsview. At the data selection with the curve fitting technique, we applied the same selection criteria to all the sites. Figure 2 shows the hourly data as well as the afternoon mean values of atmospheric CH_4 mixing ratios at the individual sites. Plotting together with the hourly data makes the three sites, highly variable. As discussed in Sect. 3.6, the influence of local-scale emission strength primarily manifests in the diurnal variations of atmospheric mixing ratios. In general, afternoon mean mixing ratios represent the source strengths over a large-scale area, exhibiting synoptic-scale events in atmospheric CH_4. However, even during the daytime, local (point) sources can interact with local-scale atmospheric condition, leading abrupt or sporadic spikes in observed atmospheric CH_4. Most of these observations of high mixing ratios are removed as outliers through this selection.

4. *167: Five days is quite short time for calculating backward trajectories. See e.g. Wittig et al.: The 10-d transport backwards in time in FLEXPART is much smaller than the average residence time of air masses (typically few weeks) in the Arctic. Therefore, part of the influence of Arctic fluxes on observations can be diluted in the background. On the other hand, backward simulations over several weeks would require a very large number of particles to be accurate, at the expense of very high computational costs. Thus, I would suggest doing a test with at least 10-d trajectories.*

The duration of the backward trajectories depends on the spatial domain of interest. Wittig et al. (2023) and our study have distinctive differences in scope and inversion setting.

We focused on Canada's CH_4 flux estimation, using the synoptic-scale variations of daily observations. The majority of footprint information, >80%, used to optimize Canada's CH_4 fluxes comes from the first three days on the course of the back trajectory (Figure R1a, in our responses to the referee #1). Wittig et al. (2023) used monthly observational data to estimate the CH_4 budget in the Arctic countries, including Canada. Their Figure 8 illustrates that less than around 10 % of the observational information is used to estimate the fluxes, while 60-75% to constrain the background atmospheric CH_4 and the rest is lost in noise.

Figure 7 in Wittig et al. (2023) shows that their inversion is most sensitive to the observations in coastal sites in and around the Arctic Circle, including Alert, Canada. On the other hand, we did not use Alert because it is far north of the continental land sources and thus less influenced by the CH_4 fluxes in Canada.

We conducted experiments to see how a 10-day footprint would affect the flux estimations. The choice of the back trajectory duration slightly changed the modelled prior mixing ratio, which was comparable to ~10% of the uncertainty with the transport error from the different transport models used in this study (see Fig. R1b in our responses to Referee #1). The difference in the estimated fluxes between 5- and 10-day back trajectories is not significant, less than 5 % for regional fluxes. Thus, we concluded that 5-day back trajectories are sufficient in the scope of our study.

5. 205: It would be helpful to see the time series or all priors, to ensure that there are no inconsistencies during the time period studied (as there might be in EDGAR time series) that might affect the trend calculation.

For anthropogenic, ECCC-AQ2013 (ECAQ) is monthly and non-varying interannually, which was repeatedly used for the entire study period. EDGAR v4.3.2 (EDGAR) covers up to 2012. After 2012, we used the same emission field for 2012 (Fig. R2). There is a slight downward trend in EDGAR in the early period, but the change is relatively small compared to the interannually varying Wetland CH_4 emissions, such as WetCHARTs and CLASSIC (also see Figure S3).

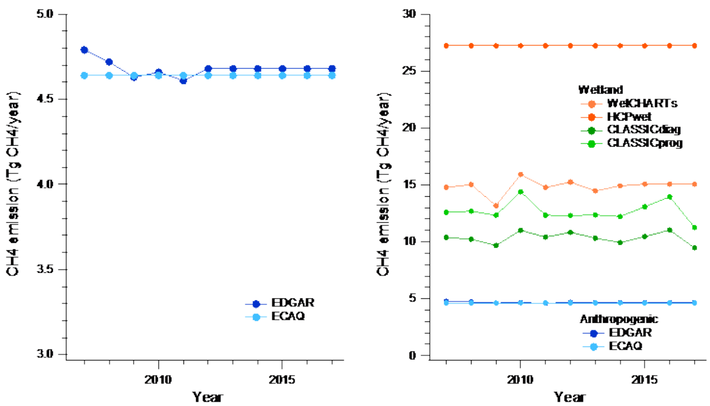

Figure R2. Time-series of prior annual CH_4 emissions. anthropogenic sources (left) and together with wetland sources (right).

As seen in Figs S5 and S6, the choice of anthropogenic emissions results in differences of the absolute values of the estimated CH_4 emission in Western Canada, but not the trends. Such differences are smaller than the spread/uncertainty of the posterior fluxes. The difference of posterior emissions in West seems to be related to the difference in the spatial distribution of the two prior anthropogenic emissions (see Fig. 3b).

We have added the time serieses of prior means to Fig. 7 as below. It would help to see there are no apparent correlation with the trend between the priors and the posterior fluxes.

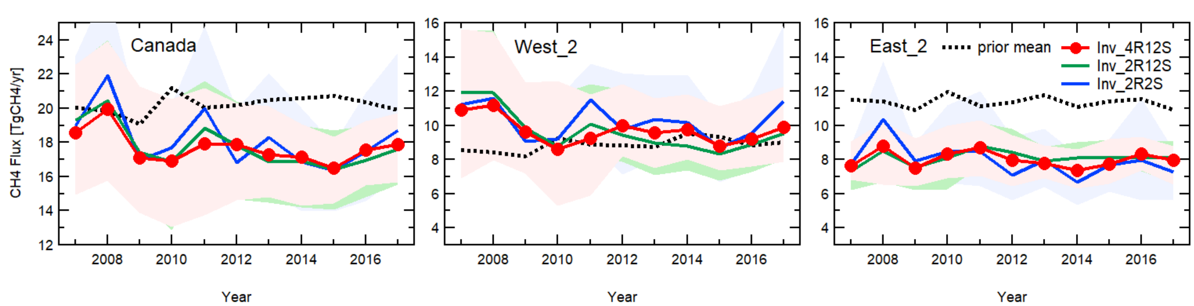

Figure 7. Trend of estimated yearly CH_4 fluxes in Canada and western (West_2) and eastern (East_2) subregions from three inversion setups, 72 experiments in total. Lines show mean fluxes over each of three inversion sets with different subregion masks and observation site selections. The shaded areas indicate the range of maximum and minimum estimates among 24 experiments per inversion setup. Black dotted lines indicate mean prior emissions.

6. *217: How do you re-grid the coarser resolution data on e.g wetland extent for use in higher resolution inversions?*

CLASSIC wetland CH_4 flux data are on a grid of 2.81°, which is only the coarser resolution than 1°×1°. First, we divided the coarse grids into small parts of 0.01° × 0.01° and then sum-up the small tiles over a grid of 1°×1° (see example below).

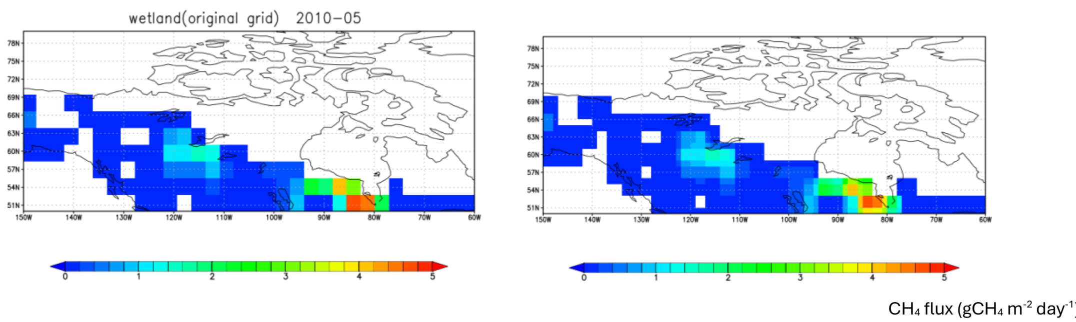

Figure R3. Example of re-griding of CLASSIC wetland CH_4 data. Original emission map at a resolution of 2.81° (left) and re-gridded 1°×1° emission map.

7. *273: More recent EDGAR releases include an annual cycle for the anthropogenic emissions. How would this affect your results?*

The impact of the annual cycle of anthropogenic emissions on the flux estimates is negligible in this study.

We did not use the seasonal varying EDGAR emissions. Instead, ECAQ includes larger seasonal variations than the seasonal EDGAR, especially in CH_4 emissions from Landfill sector. However, the seasonal variation of ECAQ anthropogenic emission is less visible, compared to the one in wetland CH_4 emissions as Figs. 3e and 3f in the manuscript. In our posterior fluxes, no clear seasonality due to the seasonally varying ECAQ was found.

8. *291: In reality, cold months can vary from year to another and the shoulder seasons may have a significant impact on methane emissions. How would this affect your results?*

There are year-to-year changes in the seasonal variations of posterior fluxes. In this study, we analyse the mean seasonal cycle over the study period. In this way, inter-annual variations are minimized in our analysis.

9. *Figure6: What could be the midwinter peak in methane emissions (especially 2011 and 2013 in East)? Anthropogenic or natural emissions?*

Figure R4 shows the year-to-year variations in the January footprints for Fraserdale. In 2011 and 2013, there appears to be more footprint influence from the province of Manitoba (with anthropogenic emissions from cities like Winnipeg) on the western end of the sub-region East. Thus, the notable winter methane emission peaks in East for 2011 and 2013 could be a combination of natural and anthropogenic emissions. In this study, the flux partitioning into natural and anthropogenic is based on the mean seasonal CH_4 fluxes. More analysis is needed to understand the inter-annual variations in the flux partitioning in the site observations.

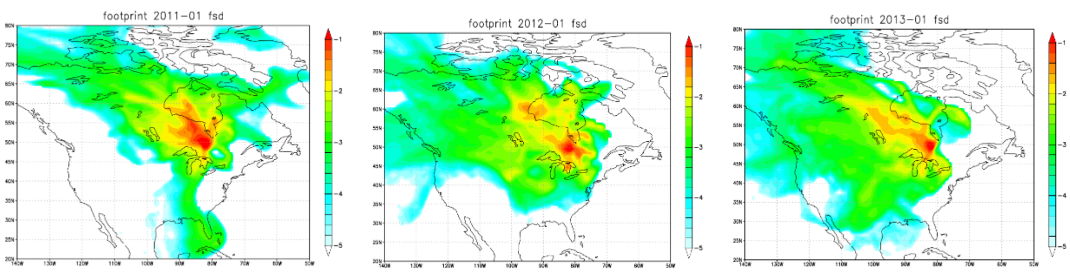

Figure R4. Monthly mean footprints in January 2011 to 2013 (left to right) for Fraserdale (FSD). Unit is $\log_{10}$ ppm/(mol /(m^2 s)).

10. *Table S2: Can you give a statistical estimate of the significance of the trend, in addition to SD among the ensembles?*

We have added p-value of the trend in Table S2 as follows:

Table S2. Ensemble mean trends of estimated yearly CH_4 fluxes, ~~and~~ uncertainties (SD among the ensembles) and *p*-values for Canada and western (West_2) and eastern (East_2) regions from three inversion setups, 72 experiments in total and 24 experiments in each of three inversion setups (Inv_4R12S, Inv_2R12S and Inv_2R2S) with different subregion masks and observation site selections. The trends are calculated as slopes of linear fit over three periods: the whole (2007–2017), the early (2007–2011) and the later (2012–2017) periods.

	Canada			West_2			East_2		
	2007–2017	2007–2011	2012–2017	2007–2017	2007–2011	2012–2017	2007–2017	2007–2011	2012–2017
Total	-0.20 ± 0.14 $p = 0.08$	-0.36 ± 0.59 $p = 0.09$	0.05 ± 0.23 $p = 0.64$	-0.15 ±0.14 $p = 0.53$	-0.49 ± 0.48 $p = 0.23$	0.02 ± 0.17 $p = 0.64$	-0.05 ± 0.10 $p = 0.77$	0.02 ± 0.27 $p = 0.92$	0.03 ± 0.12 $p = 0.65$
Inv_4R12S	-0.14 ± 0.18 $p = 0.13$	-0.42 ± 0.68 $p = 0.13$	0.00 ± 0.19 $p = 0.02$	-0.12 ±0.14 $p = 0.34$	-0.59 ± 0.53 $p = 0.07$	-0.07 ± 0.14 $p = 0.46$	-0.02 ± 0.06 $p = 0.98$	0.16 ± 0.22 $p = 0.61$	-0.02 ± 0.04 $p = 0.48$
Inv_2R12S	-0.25 ± 0.13 $p = 0.03$	-0.44 ± 0.62 $p = 0.01$	-0.04 ± 0.22 $p = 0.53$	-0.27 ±0.13 $p = 0.40$	-0.70 ± 0.47 $p = 0.11$	-0.01 ± 0.15 $p = 0.24$	0.03 ± 0.06 $p = 0.79$	0.25 ± 0.22 $p = 0.95$	-0.03 ± 0.10 $p = 0.51$
Inv_2R2S	-0.22 ± 0.08 $p = 0.17$	-0.22 ± 0.43 $p = 0.52$	0.18 ± 0.23 $p = 0.10$	-0.07 ±0.05 $p = 0.77$	-0.18 ± 0.26 $p = 0.72$	0.12 ± 0.16 $p = 0.91$	-0.15 ± 0.08 $p = 0.47$	-0.04 ± 0.29 $p = 0..63$	0.06 ± 0.11 $p = 0.58$

-0.5 -0.3 -0.1 0.1 0.3 0.5 (Tg CH_4 year^{-2})

11. 396: Could you use trajectories to select those time periods when air masses were transported directly from one site to the other?

We could not make such selection. The wind direction is always changeable. The air is diffusive over time. Because of these characteristics of the air, the likelihood of an air mass transported directly from one site to another is low. Therefore, we compared the multi-year mean concentration difference of the two sites (representing the mean transport) by month, without any selection based on wind direction/trajectories.

12. 465: How about the increase in the depth of the permafrost thaw layer, which progresses through the summer and increases the temperature of the subsurface layers? Could it, in part, explain the later emission maximum?

It could be, but not sure. In the Arctic, the seasonally thawed active permafrost layer increases in depth during the summer and starts freezing in the late August or September. The thawed permafrost increases the soil moisture, providing a favourable environment for the microbial CH_4 production. In the mid-latitude subregions, West and East in this study, there are some small areas of discontinuous permafrost layers, and sporadic or isolated permafrost patches in the deep soil (e.g., Tarnocai, et al. 2009). This type of permafrost may not be actively impact on the seasonality of Wetland CH_4 emissions.

13. *694: Could it be possible that the diurnal cycle may also be influenced by anthropogenic sources, as the LLC was described as having oil industry in the south of the site.*

From this context, we assume that LLC is meant to be LLB (Lac La Biche). ECCC has another monitoring station near LLB (~400 –500 km away) at Esther (EST) in CHOPS (Cold Heavy Oil Production with Sand) region with known local anthropogenic sources. Figure R5 show the seasonal cycles of normalized diurnal amplitude at EST and SD along with LLB. On average, winter diurnal amplitudes at both sites are similar. However, the amplitude at EST is lower than SD. This result indicates that the strength and spatial distribution of local CH_4 sources around EST are irregular in time as the local anthropogenic CH_4 sources are fugitive from the oil/gas industry. In contrast to EST, the diurnal amplitude at LLB is more prominent than SD around the year. This distinctive difference between LLB and EST supports the existence of regular CH_4 sources around the LLB; that is, they are presumably natural sources.

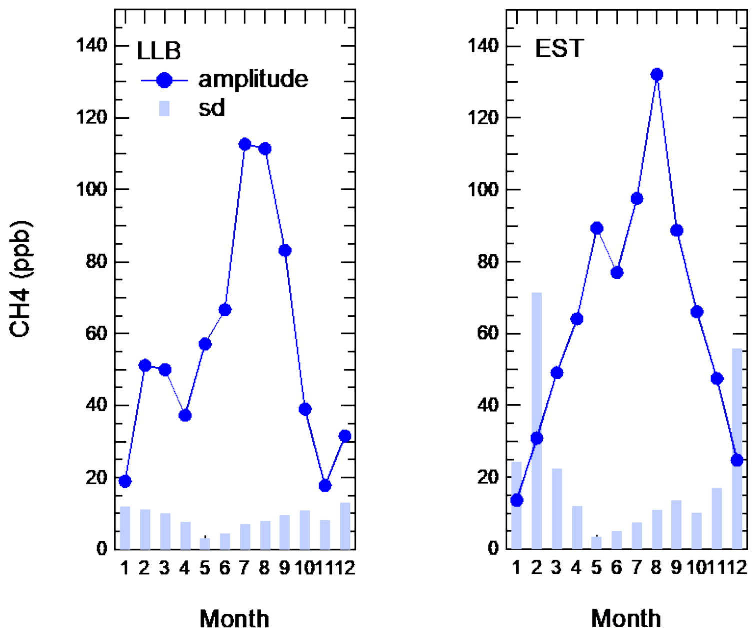

Figure R5. Seasonal cycle of normalized diurnal amplitude and SD of observed atmospheric CH_4 during the afternoon mean (14–16 local time based of normalization) for Lac La Biche (LLB, same as shown in Fig 14), and Esther (EST).

References

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