Reply to Comments by Referee #2

We thank the referee for constructive and helpful comments to improve our manuscript with more clarifications. We address the comments (italic and red). In the responses, we also indicate the changes made in the manuscript (in blue font).

General comments

This manuscript presents atmospheric observations of CH4 from 5 new in-situ measurements sites in the Canadian Arctic and uses these (plus one other site) in atmospheric inversions to determine the land-atmosphere flux of CH4. The authors find notable inter-annual variability in the natural (wetland) flux, which may be related to variations in surface temperature. Although the study is interesting and fairly well presented, further explanations and clarifications for some of the methods are needed before being published. In addition, minor technical corrections for English language usage are required.

We report the modifications and additional explanations that we made in the manuscript.

Specific comments

P1, L14: I suggest specifying the number, instead of "multiple". Also how is "inversion modelling system" defined, by the inversion algorithm or transport model used? In this study 2 different transport models were used with 3 different meteorological datasets, so I suggest the authors state this instead.

As suggested, we have changed the sentence more specifically:

From:

Multiple regional Bayesian inversion modelling systems are applied..

To:

Three regional Bayesian inversion modelling systems with two Lagrangian Particle Dispersion Models and three meteorological datasets are applied…

P1, L30: I suggest the authors state what the carbon is vulnerable to, i.e., conversion to CH4 and CO2 which can be emitted to the atmosphere

After "vulnerable", we have added "to conversion to CH_4 and CO_2 which can be emitted to the atmosphere".

P2, L5: Please specify the magnitude of what, presumably CH4 emission but this should be stated

Yes, it is the magnitude of CH_4 emission. We have changed to:

"… show large discrepancies in the spatial distribution of wetland CH_4 source, as well as its magnitude"

P3, L7 (and throughout): It's actually CH4 volume mixing ratio that is reported, and not concentration, so I suggest changing "concentration" to "mixing ratio" throughout.

As the referee noted, gaseous concentration is frequently referred to as 'volume mixing ratio'. We have changed from "concentration" to "mixing ratio", throughout the manuscript.

P5, L31: By "SD of the observed time series to their fitted curves" do the authors mean the SD of the residuals, i.e., after subtracting the fitted curves? This is not clear.

Yes, we mean SD (standard deviation) of the residual of the observations from a fitted curve. That is also referred residual standard deviation. For clarification, we have added "residual" into the sentence:

"monthly Standard Deviation (SD) of the residual of observed time series..."

P5, L34 to P6, L2: This needs some explanation why the difference between SD_PM and SD_24 gives an indication of whether the daily variability is due to local scale changes in emissions or seasonally changing atmospheric transport. I guess the authors mean that SD_24, which includes also night-time data, is more sensitive to local emissions than SD_PM, but an explanation should be provided.

We use the difference between SD_PM and SD_24 as an indication of local emission. SD_24 includes nighttime data which is more sensitive to local emissions than SD_PM. For clarification, we have added the following sentences:

The nighttime planetary boundary layer (PBL) is usually shallow, while the daytime boundary layer is usually deeper and well mixed. If there are local CH_4 sources, the emission is mixed into a shallow PBL at night (yielding higher mixing ratio) and deeper PBL during the day (yielding lower mixing ratio). The resultant diurnal variations in the CH_4 mixing ratios are evident as larger CH_4 SD_24 compared to SD_PM. In the absence of local sources, SD_24 is comparable to SD_PM.

P6, L6: "rectified" is not the right term here (the rectifier effect is a specific term given to the co-variation of flux and planetary boundary layer height, particularly for CO2, which doesn't apply here). Instead use "amplified".

As the referee suggested, "amplified" is more suitable than "rectified". We have revised the text accordingly.

P10, L7: The authors should change this sentence to either "Our Bayesian inversion optimizes…" or "The Bayesian inversion used here optimizes…" to make it clear that the approach used here is not the only approach.

We have changed:

From: "The Bayesian inversion optimises…"

To : "The Bayesian inversion used here optimises…"

*P10, L16: The authors state that the matrix **K** is the product of **M** (the footprints) and **x** (the surface fluxes) and is a Jacobian matrix of flux sensitivities. The elements of **K** must be in mass mixing ratio units (i.e. the same units as **y**), so by definition this is not a Jacobian matrix (but **M** is a Jacobian). Also, the dimensions of **M** and **x** should be given.*

As the referee noted, our usage of 'Jacobian matrix of flux sensitivities' is not correct. To avoid confusion, we have removed the term-"Jacobian matrix" from the sentence and revised with the addition of the dimensions of $\boldsymbol{M}$ and $\boldsymbol{x}$. The text has been changed:

From:

$\boldsymbol{K}$ is the matrix of contributions from R sub-regions. $\boldsymbol{K}$ is a Jacobian matrix of flux sensitivity, a product of two matrices, $\boldsymbol{M}$ and $\boldsymbol{x}$. $\boldsymbol{M}$ is the modelled transport (or footprints in this study), and $\boldsymbol{x}$ is the spatial distribution of the surface fluxes.

To :

$\boldsymbol{K}$ (N×R) is the matrix of contributions on the observations (N) from all the fluxes (R) of sub-regions. $\boldsymbol{K}$ is a product of two matrices, $\boldsymbol{M}$(N × LL) and $\boldsymbol{x}$ (LL×R), $\boldsymbol{M}$ is the modelled transport (or footprints in this study), and $\boldsymbol{x}$ is the spatial distribution of the surface fluxes. LL (=LAT×LON) is the dimension of our domain (1°x1° in latitudes (LAT) by longitudes (LON)).

P10, L22: The units of the observation uncertainty should be specified, presumably this is ppb. Also, an explanation should be given of how the value of 0.33 was derived, especially as this seems rather small. Furthermore, an estimate of the appropriateness of the uncertainty estimates should be given, e.g. the value of the reduced-chi-square statistic.

The 33% (0.33) prior model-data mismatch is comparable to other regional inversion studies (e.g. Gerbig et al. (2003), Zhao et al. (2009)). Zhao et al. (2009) included uncertainties from LPDM dispersion, wind field, aggregation and background mixing ratio to estimate prior model-data mismatch uncertainty. However such estimate has many assumptions that are difficult to evaluate. In this study, we tested the sensitivity of the inversion results to this setting by using 33% and 66%, as the model-data mismatch errors. The posterior fluxes changed by less than 5% for all sub-regions (and the different sub-region masks), indicating that the flux estimates were not highly sensitive to the prior error specification.

In response to a similar comment from referee 1, we have added more details (in blue below) to clarify page 10, lines 23-24:

'We examined the inversion's sensitivity to these uncertainties by doubling their values. The posterior fluxes changed by less than 5% for all sub-regions (and the different sub-region masks). The results showed the optimised fluxes are not strongly dependent on these prescribed uncertainties.'

In statistical error analysis, the reduced chi-square test qualitatively measures the goodness of fit of the model to the observations (Hughes and Hase, 2010, Drosg (2009)). In the limit of infinite number of data points and the data are independent and normally distributed, the value of reduced chi-square should be 1. Following Drosg (2009), the reduced chi-square is given by:

$$reduced\ \chi^2 = \frac{1}{N}\sum_k \sum_j {\chi_{jk}}^2$$

Where chi-square is:

$${\chi_{jk}}^2 = \frac{\sum_i \left(model_{ijk} - obs_{ijk}\right)^2}{{\sigma_{jk}}^2}$$

$$obs = observation - model_{background}$$

$$\sigma_{jk} = standard\ deviation\ \ of\ observation\ for\ month\ j\ at\ site\ k$$

$$i = 1, n_{jk}\ (number\ of\ observation\ for\ month\ j\ at\ site\ k)$$

$$The\ estimsted\ degrees\ of freedom\ N:$$

$$N = \left(\sum_k \sum_j n_{jk}\right) - (fluxes\ per\ region \times number\ of\ region \times number\ of month)$$

The overall reduced chi-squares for our experiments are:

	Mask A	Mask B	Mask C
	YT, NT, NU	YT+NT, NU	YT+NT+NU
FLEXPART_EI	1.244	1.237	1.262
FLEXPART_JRA55	1.236	1.234	1.245
WRF-STILT	1.255	1.249	1.266

Given that the observations are not normally distributed (more frequent high and very high mixing ratio events than low mixing ratio events) and the limited amount of observations, there does not seem to be a strong reason to reject the model results.

We have added a paragraph on the assessment (evaluation) of our model results with reduced chi-square statistics in Section 4.6 [Comparison of modelled and observed mixing ratios (formerly Section 4.5. Comparison of prior and posterior concentrations to observations)]:

Another qualitative measure of the goodness of fit of the model to the observations is the reduced chi-square statistics (Drosg., M., 2009;. Hughes, I. G. and T. Hase, 2010). In the limit of infinite number of data points and the data are independent and normally distributed, the value of reduced chi-square should be 1. The overall reduced chi-squares for all our experiments are in a narrow range of 1.23—1.27. Given that the observations and modelled mixing ratios are not normally distributed (more frequent high and very high mixing ratio events than low mixing ratio events) and the limited amount of observations, there does not seem to be a strong reason to reject the model results

P10, L29-30: This needs a bit more explanation, do the authors mean that they have separate variables for the biomass burning and other emissions, which are optimized simultaneously. In this case, the total number of variables would be R x 2 x number of flux time steps.

Yes, R should be the number of all fluxes to be solved. We solved two fluxes (biomass burning and other missions) per sub-region. We have changed the sentence (note: originally on page 10, lines 14-15):

From : R is the number of sub-regions to be solved.

To : R is the number of fluxes to be solved. R is two fluxes per sub-region × number of sub-regions (i.e., 2 to 6 in this study).

Section 3.3.2: Using only 3 regions for the optimization represents a significant aggregation error, as it is assumed that both the spatial pattern and relative magnitudes of the fluxes within each region are correct. Why was the inversion performed only for these coarse regions? Other than being different territories, are they characterized by having similar ecosystems, climate or other?

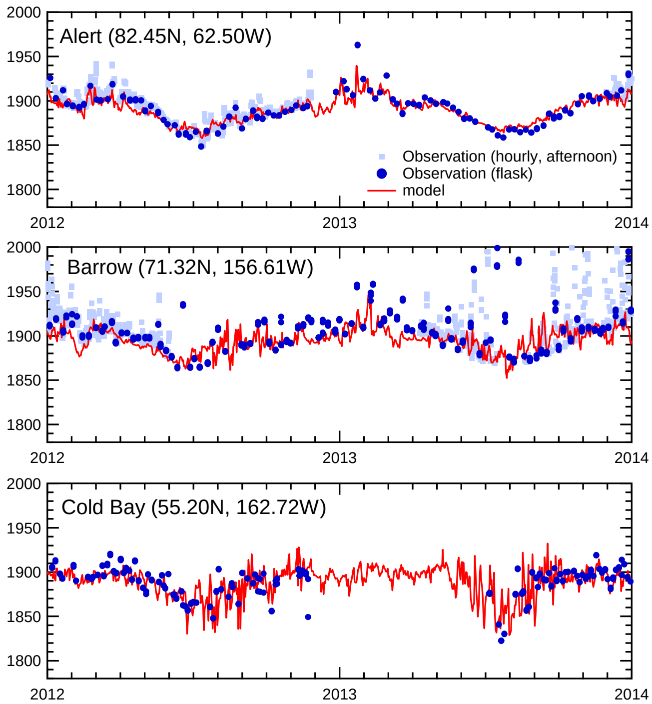
We tried to account for the potential errors in the spatial pattern and relative magnitudes of the fluxes by using three different priors to provide a range of spatial and flux magnitude patterns. In our results, the prior flux error is smaller than the model transport error (as estimated by the different transport models used in this study).

The number of sub-regions that could be resolved by the inversion depends mainly on the amount of observations (spatial coverage density and strength of regional signals above the background variations) and magnitudes of the transport errors. In the absence of transport errors, the inversion can resolve a large number of sub-regions (an order of magnitude more in some experiments we tried). But with the present transport model errors, we begin to see unrealistic (negative fluxes) in weak flux region (Yukon) sporadically in our results. Hence, observations and model errors limit the number of sub-regions used in the inversion. The regions were defined based mainly on the geographical characteristics. Yukon has many mountains and little wetlands. Northwest Territories is mainly lowlands with most of the wetland in the Canadian Arctic. Nunavut is a part of the Canadian Shield or Laurentian Plateau with limited wetlands.

Section 3.3.3: Errors in the modelled background can incur errors in the posterior fluxes. Did the authors check their global modelled CH4 mixing ratios (from NIESTM) with independent observations in the northern high latitudes? Was an estimate of the uncertainty in the background made and included in the overall observation uncertainty?

Yes, we checked the performance of NIES-TM with the independent observations. Here, we showed the comparison of model and observations for the sites, Alert, Barrow and Cold Bay, which were not used for the inversion in this study. Overall, the model captures the observed variations at synoptic scale to seasonal and long-term trend.

Yes, in our inversion, background uncertainty is implicitly included in observation error as we mentioned in the earlier response.



P12, L21: The sub-region masks A to C are not defined in the text

The sub-regions are defined in the earlier section 3.3.2. But the masks A, B and C have not been stated in the text, thought they are illustrated in Fig. S4. The sentence in Section 3.3.2 have been changed:

From:

We set up three sub-region masks for the Canadian Arctic based on three territories 1) Northwest Territories (NT), Yukon (YT), and Nunavut (NU), as shown in Fig. S4

To:

For the Canadian Arctic based on three territories, Northwest Territories (NT), Yukon (YT), and Nunavut (NU), we set up three sub-region masks, Mask A, B and C, as shown in Fig. S4.

P12, L26: I suggest the authors state that the negative biomass burning fluxes are "spurious" since the biomass burning source cannot be negative.

We have modified by adding a sentence as suggested:

As a result, negative mean fluxes, i.e. CH_4 sinks, could appear, especially in YT (Fig. 8a); the negative biomass burning fluxes are "spurious" since the biomass burning CH_4 source cannot be negative. However, a null-flux would be consistent within error bars.

P15, L18-19: Do the authors mean the anomalies of the deseasonalised data? It is important to look at the anomalies in the data after the mean seasonality has been subtracted to avoid correlations with temperature between months, which would override possible correlations with temperature between years.

Yes, the anomalies of fluxes and meteorological parameters we discussed there are the de-seasonalised data by subtracting the 4-year averaged monthly mean values. .

Figure 12: It would be interesting to see the regressions for the prior wetland emissions as well. How strongly are the prior wetland emissions correlated with the meteorological variables and how does this influence the posterior correlations?

In our study, natural CH_4 fluxes (wetland and other fluxes except biomass burning CH_4 flux) in prior emission cases, C1 and C2, are multi-year mean monthly fluxes. Therefore they have no year-to-year anomalies and no correlation with the meteorological anomalies. Only for C3, the prior wetland CH_4 fluxes from WetCHARTs ensemble mean exhibit inter-annual variation, the correlations with temperature and precipitation anomalies are $r = 0.34$ and $r = 0.92$ respectively.

The table below shows the correlation coefficients of the natural (wetland) posterior fluxes and the meteorological variables for individual emission scenarios along with the correlation coefficients of the prior natural fluxes. The posterior natural fluxes in C3 with WetCHARTs prior fluxes show slightly higher correlations than those in the other two cases with cyclo-stationary prior fluxes. But overall there is no significant dependency of posterior correlation on the prior wetland fluxes. This result indicates that the inter-annual variations in the posterior wetland fluxes are mainly determined by the observations, rather than by the prior fluxes. Note that as following Referee 1's suggestion, we have changed C1, C2 and C3 to VIS, GEL, and WetC respectively.

	Natural			
	temperature		precipitation	
	prior	posterior	prior	posterior
C1 (VIS)	0.00	0.55	0.00	0.11
C2 (GEL)	0.00	0.54	0.00	0.07
C3 (WetC)	0.26	0.55	0.90	0.16

In the revision, we have added texts to explain the prior flux influence on the posterior flux-climate correlations in the section of "Relationship of fluxes with climate anomalies (now in Section 4.5).

In prior cases VIS and GEL, natural CH_4 fluxes (wetland and other fluxes except biomass burning CH_4 flux) are multi-year mean monthly fluxes. Therefore these prior fluxes have no year-to-year anomalies and no correlation with the meteorological anomalies. Only in WetC, the prior with wetland CH_4 fluxes from WetCHARTs ensemble mean exhibits inter-annual variations, the correlations with temperature and precipitation anomalies are r = 0.26 and r = 0.90 respectively. The posterior natural fluxes with WetC show slightly higher correlations (r= 0.55 with temperature, r=0.16 with precipitation) than the mean correlation values. But, overall there is no clear dependency of posterior correlations on the inherent climate anomaly correlations in the prior fluxes. This result indicates that the inter-annual variations in posterior wetland fluxes in this study are mainly determined by the observations, rather than by prior fluxes.

Technical comments

P1, L26: "stronger then" should be "stronger than"

Corrected.

P1, L27: add "from" before "about 722 pbb"

Added "from"

Generally: attention should be paid to the use of articles "the" and "a" and when no article should be used at all.

Thank you for your comment, we reviewed the text and tried to correct the usage of articles.

P6, L14: replace "Like" with "Similar to" as "like" in this sense is very colloquial.

Changed from "Like" to "Similar to"

P6, L15: there are words missing in this sentence, it should be "…indicates that there is a weaker local source of CH4…" and "than around the three continental sites".

Corrected, by adding the words as follows:

This indicates that there is a weaker local source of CH_4 around CBY than around the three continental sites.

P6, L19: should be "suggested that there are on-going CH4 emissions from…"

Changed:

From: suggested the CH_4 emissions from

To: suggested that there are on-going CH_4 emissions from

P6, L29: should be "due to the (very) short period of daylight"

Changed:

From: due to limited winter daytime

To: due to the short period of daylight

P8, L27: should be "C3 is the same as used in C2, but…"

Corrected.

P9, L31: should be "…map of climatological termite emissions"

changed

From: a climatological emission map of termite"

To: map of climatological termite emission"

P12, L16: change "done" to "made"

Changed.

P12, L22: should be "are shown" (not "showed")

Corrected.

Fig. 5: should be "same as C2"

Corrected.

P14, L25: Suggest changing the section heading to "Sensitivity tests" since there are more than one

As suggested, we changed the section heading:

From: Sensitivity test

To: Sensitivity tests

P15, L7: should be "in winter compared to…" (not "against") and I think the authors mean "which might contribute to large uncertainties in the flux estimation"

We have changed:

From: in winter against the observed concentrations, which might have large uncertainties in flux estimation.

To: in winter compared to the observed CH_4, which might contribute to large uncertainties in flux estimation.

P15, L9: change "done" to "made"

Changed.

References:

Drosg., M., Dealing with Uncertainties: A Guide to Error Analysis , Second Edition Manfred Drosg Springer, 2009.

Gerbig, C., J. C. Lin, S. C. Wofsy, B. C. Daube, A. E. Andrews, B. B. Stephens, P. S. Bakwin, and C. A. Grainger, Toward constraining regional-scale fluxes of CO_2 with atmospheric observations over a continent: 1. Observed spatial variability from airborne platforms, J. Geophys. Res., 108, 4756, doi:10.1029/2002JD003018, D24, 2003.

Hughes, I. G. and T. Hase, Measurements and their Uncertainties: A Practical Guide to Modern Error Analysis Oxford, 2010.

Zhao, C., A. E. Andrews, L. Bianco, J. Eluszkiewicz, A. Hirsch, C. MacDonald, T. Nehrkorn, and M. L. Fischer (2009), Atmospheric inverse estimates of methane emissions from Central California, J. Geophys. Res., 114, D16302, doi:10.1029/2008JD011671, 2009.