We thank again the reviewers for their comments. Our pointwise responses (AR) to reviewers' comments (RC: in *Italics*) and the respective changes in the manuscript (blue text) are as follows:

Reviewer #1

RC*: The authors have addressed and answered to the reviewers' comments carefully, and revised the manuscript accordingly. The manuscript is improved well, and I suggest it to be published after revising a few minor points below.*

P1 L20: "as it accounts for" -> ", accounting for"
P4 L120: "be chemically removed" -> "be reduced (or become negligible?) due to atmospheric chemistry"
P4 L126: "a unit initial mole fraction" -> "an uniform initial mole fraction"?
P7 L208: "methane. sufficient" -> "methane, sufficient"?
P7 L218: "RMSD (root mean square difference)" -> "root mean square difference (RMSD)"
P7 L222: "prior observation RMSD"
Do you mean "prior (observation-prior?) RMSD", "observation RMSD" or something else?
P8 L238: "and over TRANSCOM regions" -> "and TRANSCOM regions."
P8 L243: "for a prior of" -> "with the prior emission of"
P8 L234: Please add +/- sign before "0.5 Tg" to be consistent with other adjustment numbers.
P8 L247: "The South American temperate region has the largest difference between the serial and PP emission estimates of 2 Tg yr-1"In the previous sentence, you mention that the differences in Eurasian temperate is also 2 Tg. Please modify the sentence to make it clear what you meant to say.
P9 L280-281: Please merge with the previous paragraph. One-sentence paragraph is not appropriate.
Figure 1 caption. Please add a note that the diagram is an example when splitting the inversion into three blocks or modify the diagram to be more general with k blocks (e.g. H1, H2, H3 -> H1, H2,..Hk).
Figure 5 caption: "emission estimates of" -> "emission estimates from"
Figure 5 caption: "(see Figure 1)"
Did you really meant to refer to Figure 1 or regional definition illustrated in Figure 2?

AC: We have corrected the manuscript following the suggestions of the reviewer.

Reviewer #2

I have found the manuscript has greatly improved from the previous one. Therefore, I recommend this for publication after minor corrections suggested below.

Minor comments:
L34: "CTM's" => "CTMs"

L54-55: "The method has the advantage… non-linear inverse problems." Please note that even a variational method assumes linearity in its algorithm. Therefore, I do not agree with this statement.

AC: The variational method can be applied to a weakly non-linear inverse problem when combined with an appropriate steepest-decent numerical minimizer. For example, Pandey et al (2015, 2016), Krol et al. (2013) and Naus et al. (2021) have performed non-linear inversions using the variational method. They use the M1QN3 minimizer, based on a quasi-Newtonian algorithm, which is more suitable for non-linear inversions (Gilbert and Lemaréchal, 1989).

We have modified the sentence and listed some of these studies:

"The variational approach can be applied to weakly non-linear inverse problems using a suitable steepest-decent numerical minimizer (Naus et al., 2021; Pandey et al., 2016)"

L116: "$|x|$ denotes the global sum" I think this is very confusing because "$|.|$" is usually used for representing absolute values.

AC: We have removed $|x|$ from the equations. We now denote the summation matrix **E**.

"Here **E** denotes a summation matrix used to compute global sum of the elements of $\boldsymbol{x_l}$."

L145: "The correct adjoint implementation" => "The correctness of the adjoint implementation"
Eq. (9): How do you verify the equality? Should the equation be satisfied within round-off errors?

AC: The adjoint test checks if the equality (Equation 9) is satisfied to an accuracy near the computing precision. We have added the information to the revised manuscript.

"The test checks if the equality

$$\langle M(\boldsymbol{a}), \boldsymbol{b} \rangle = \langle \boldsymbol{a}, M^{*}(\boldsymbol{b}) \rangle \quad \ldots\ldots\ (9),$$

is satisfied to an accuracy near the computing precision"

L157: "This can be avoided by taking a …" Please elaborate how "another inversion" is prepared in advance".

AC: We have added the following text to the revised manuscript:

"This can be avoided by taking a realistic $\boldsymbol{c}_0$ from the posterior mole fractions simulations of another inversion covering the period before the PP inversion. If such an inversion is not available, $\boldsymbol{c}_0$ can be computed by performing an inversion for the 1-year period preceding the PP inversion."

L179-182: I have a concern about transport biases induced by a different resolution.
If you changed the model resolution, some bias (persistent difference from the original resolution) could arise (e.g., exchange rate between the upper-troposphere and the lower-stratosphere).

AC: We have removed this text from the revised manuscript to address the issue raised by the reviewer.

L208: "methane. sufficient for our test inversion" typo?

AC: We have corrected the typo.

3.2 Wall time: Could you specify the CPU you used here?

AC: We used "12-core 2.6 GHz Intel Xeon E5-2690 v3".

L281: "conventional" may be better than "traditional"
Also, I'm wondering if "11-year" is traditional…

AC: We have changed the text to avoid confusion.

L283-289: Please consider to make this paragraph to an independent subsection (i.e. 4.1).

AC: Done.

Reviewer #3

The authors present an implementation of a so-called physical parallelization (PP) for variational flux inversions: from a previously described PP aimed at carbon dioxide (CO2), they add developments to take into account the chemical reactivity of methane (CH4).

General comments

As already stated in the review of the first version of the paper, the developments described in this paper are particularly relevant since long-term methane inversions are now run by several teams and the issue of the trends in methane emissions by various types of sources is still under study. The revisions made after the first review have lead to a clearer introduction and description of the work. Technically, the notations are now clearer in the mathematical description. The writing has been improved but I think, although I am not an native English speaker, that some small mistakes remain.

The introduction to the paper is now clear and focuses on the main relevant points so that the reader understands why this implementation of PP is interesting for methane inversions at the global scale.

In Section 2 Physical parallelization for methane inversions, it is now clear which parts are the general or Chevallier (2013) developments and which are specific to this work and therefore, to methane.

In Section 3 PP Performance test, only the prior uncertainties are now used, which makes the message simpler.

Section 4 Discussion contains elements which have been asked for by the reviewers of the first version and makes clear the potential of the PP.

Specific comments

Section 1 Introduction
- p.2 l.36: "Inversions have been performed on multidecadal scales to assess the information content of long records of methane mole fractions." This sentence is a bit strange to me because running an inversion means that it is assumed that the assimilated data does actually contain information on the fluxes (and other variables). It would be good to be able to assess the information content prior to running inversions, which is sometimes approximated by using sensitivity studies.

AC: We have modified the sentence:

"A few studies have performed inversions on multidecadal scales to constrain emissions using the long measurement record of methane mole fractions."

Section 2 Physical parallelization for methane inversions
- p.3 l.81: the choice of x^a for the prior is not very good because in the notations used for analytical inversions, ^a denotes the posterior. Usually, the prior state vector is noted x^b.

AC: We have replaced x^a with x^b.

- p.4 l.108: "the emission differences between the prior and the iteration in the period preceding the block": it is not very clear what is preceding what: is it the same iteration but for the preceding period of time? the same period of time but the previous iteration? the previous iteration in the preceding period of time?

AC: We have modified the text:

"Here the scalar n_k^i accounts for the global mean mole fraction changes due to emission differences $(\boldsymbol{x}^i - \boldsymbol{x}^b)$ during the inversion period that precedes the block k."

- p.4 l.114 Eq. 5: the [and] do not seem to be necessary, they give the idea that f is a function of x - x^a.

AC: Following a suggestion of the 2nd reviewer, we have removed the |x | notation. We need to use "[ ]" to denote precedence of operations in the updated equation. We denote functions using "()", for example, in Equation 4.

- p.4 l.123-124: "implementing an atmospheric sink operator S": here, either give more details on what is in this operator or put a reference to the section which contains the information.

AC: Done

- p.4 l.122: Fig. 1 is not totally clear and consistent with the text here. The colours red and green in Fig. 1 are not explained in its legend so that a first idea of the reader is that what is in red comes from Chevallier (2013) and what is in green is linked to this work.

But the last sentence of the legend states that it is the blue boxes which indicate which part comes from which work. Nevertheless, the "corrections calculation" is from Chevallier (2013) according to Fig. 1 but from this work Please make the text and figure very clear on which parts come from whose work.

AC: The "corrections calculation" step was implemented by Chevallier (2013) as "global mass increment" (see Equation 4).

We have modified the text to clear other confusions:

"In an iteration, the block mole fractions for the iteration $\boldsymbol{m}_k^i$ is computed using the block CTM operator H_k, the iteration emissions for the block $\boldsymbol{x}_k^i$, the initial mole fraction for this block $\boldsymbol{c}_k^b$, and a mole fraction correction n_k^i"

We have added the following to the legend of Figure 1:
"The steps shown with red boxes use CTM runs and take long wall time. The steps shown in green are without CTM runs and require negligible wall time

- p.5 l.131: "Sl,k accounts for the impact of atmospheric sinks": the reader still does not know at this point what is in S and the link to Sl,k. A paragraph dedicated to the sink, which is the main feature of the PP elaborated here is required at some point.

AC: We clarified and added more information on the *S*:

"$\boldsymbol{n}_k^i = \boldsymbol{h}_k \sum_{l=1}^{k-1} s_{k,l}\ f\ \boldsymbol{E}\ [\boldsymbol{x}_l^i - \boldsymbol{x}_l^b]$ ....(6).

Here the scalar $s_{k,l}$ accounts for the impact of atmospheric sinks on the global uniform mole fraction change during the time period between the blocks k and l. $s_{k,l}$ is generated using a sink operator S. We describe a formulation of S in the next section."

"We parameterize the sink operator *S*, which computes the sink scaling factor $s_{k,l}$ (Equation 6), with an e-folding decay function and a constant atmospheric lifetime of methane (τ) of 9 years.

$s_{k,l} = S(k,l) = e^{-|t_l - t_k|/\tau}$ .... (10)

Here t_l and t_k are the start times of the blocks *l* and *k*, respectively. We found this simple parameterization with a constant lifetime is sufficient for our test inversion. "

*- p.5 l.143 Eq 8: the notations are not clear to me in this equation. The adjoint of S is denoted by *, which indicates that it is not assumed to be linear. But the adjoint of h is indicated by T, which indicates that it is linear. This is not totally consistent with H begin the CTM operator, with an adjoint denoted by * in Eq. 3 and p.5 l.138 above. Could you provide more details on the derivation of the equations and make the notations totally consistent?*

AC: For *S* operator, see our response to the previous comment. We have clarified $\boldsymbol{h}_l^{\mathrm{T}}$ in the revised manuscript

"$g_k^i = f \sum_{l=k+1}^{r} s_{k,l}\ \boldsymbol{h}_l^{\mathrm{T}}\ \delta\boldsymbol{m}_l^i$ (8),

Here $\boldsymbol{h}_l^{\mathrm{T}}\ \delta\boldsymbol{m}_l^i$ is the matrix dot product of the two vectors, both of which have the same size"

- p.6 l.161 seq.: the summary of the practical steps of the PP is a good idea but there is nothing about the sink. Please add the necessary steps since it is the main feature of the PP described here.

AC: We have added the additional step to the summary.

"Prepare a sink operator *S* which accounts for the impact of atmospheric sinks on methane mole fractions during a period."

Section 3 PP Performance test

- p.7 l.215: the title of the subsection is "Emission estimation errors" but the first two paragraphs are about the match in the concentration space. Maybe it would be clearer to make a subsection dedicated to the concentrations separated from the subsection on the emissions.

AC: Done.

- p.8 l.228: "For both inversions, a good model fit to the observations" Shouldn't "a good fit" be defined, if possible with reference to R?

AC: We have now defined a good fit as "90 % reduction in mean of observation-model mismatch".

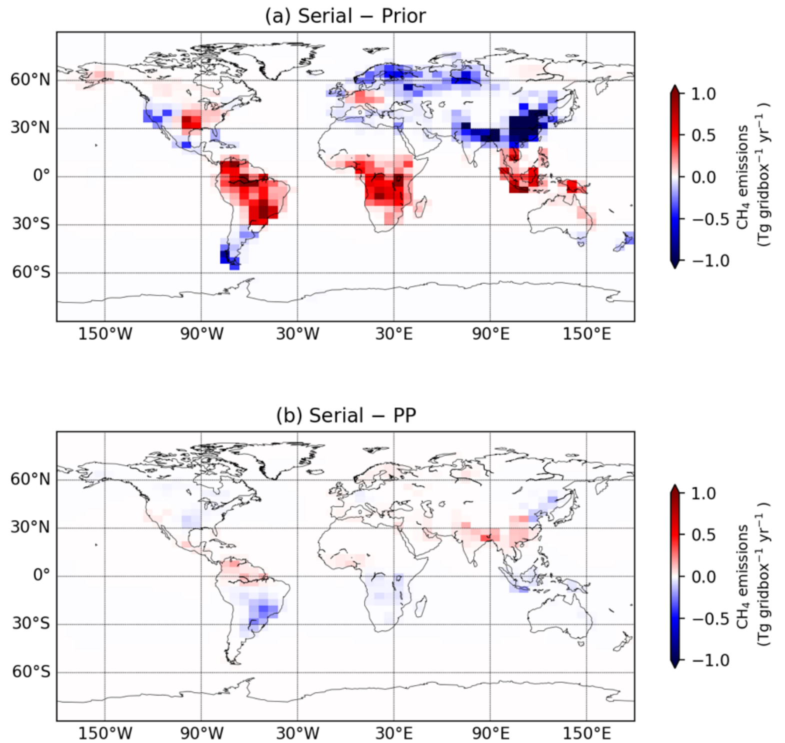
- p.8 l.237: "should be in good agreement" Same remark as above: how is a "good" agreement defined?

AC: We have quantitively described the PP errors in the Section 3.1.2. We have modified the sentence to clarify:

"The physically parallelized CTM used in the PP inversion has lost some of the consistency of the full CTM used in the serial inversion and the PP emission errors will depend on the impact of this CTM simplification."

- p.8 l.237-238: "from the inversions integrated over the globe and over TRANSCOM regions". According to the description of B (p.7 l.195 seq.), the inversions are performed at the pixel's resolution. It is a bit strange therefore to assess the performances of the PP using (very) large regions. I guess a lot of inversions can be in "good" agreement over the whole globe since the constraint on the mean total emissions of methane is strong (as stated l.240-241 "The global methane emissions are in general well constrained by the NOAA observations in a serial inversion"). The same applies for large regions such as TRANSCOM's: for example, an inversion with homogeneous emissions inside a region and an inversion with a dipole of negative/positive increments can give the same total over a large region. I understand that it is not simple to compare two inversions at the grid cell's resolution but showing maps would seem to me to be more relevant than the average global total.

AC: Inversion studies of methane (and CO_2) assess emissions at regional scales because of the reasons that the reviewer has stated. Also, we use NOAA background observation in our test inversions, which does not provide a good constraint on the grid scale emissions. Although, we believe that the assessment of the emissions as global and TRANSCOM regions total is more relevant than grid cell resolution, we have added a figure to the manuscript showing the emission differences at grid scale following the suggestion of the reviewer:



"**Figure 8 Figure 8** Emission differences averaged over 1999-2010. Panel (a) shows differences between serial and prior. Panel (b) shows differences between serial and PP"

AC: We have added the following text to the Section 3.2.1:

"Figure 8 shows the spatial distribution of the emission differences at grid scale. The mean (± 1σ spread) of the differences between the serial inversion and prior is -8×10^{-3} ($\pm$ 0.5) Tg gridbox^{-1} yr^{-1}, and it is 9×10^{-5} ($\pm$ 0.04) Tg gridbox^{-1} yr^{-1} for serial and PP inversions. Emission differences between the PP and serial inversions are visible over India and South American temperate. These differences are likely due to the lack of observational constraint in these regions (see Figure 2)."

- p.8 l.241-242: "the additional error introduced by the PP method does not seem to have a significant impact on the global emissions': please define "significant".

AC: We have made the sentence more quantitative:

"The global methane emissions are in general well constrained by the NOAA observations in a serial inversion, and the additional error introduced by the PP method only causes a 1 % error relative to the serial-prior emission mismatch."

- p.8 l.244: "is well within the prior emission uncertainty". I don't understand the argument here. The difference between the two inversions should be such that they are inside each other's (posterior) uncertainty; the prior uncertainty is what the inversion aims at reducing so it does not seem to be a good measure for a "small" difference.

AC: We do not have a good estimate of the posterior uncertainties. Therefore, we evaluate the PP-serial emission differences relative to prior uncertainties. We have added the following text to clarify:

"We do not have a good estimate of the posterior uncertainties because a large number of variational inversion iterations are needed for the second derivative of the cost function to converge. Therefore, we evaluate PP performance by comparing the PP-serial emission differences against the emission adjustments performed by the serial inversion (serial-prior differences) and prior emission uncertainties."

- p.8 l.249: "estimates for the TRANSCOM regions deviate within < 5 % from the serial emissions": how can you convince the reader that 5% is a small difference?

AC: We have modified this sentence in the revised manuscript as follows:

"In summary, mean PP emission estimates for the TRANSCOM regions deviate within < 5 % from the prior emissions, while the serial-prior differences are up to 50 % of the prior emissions."

- p.9 l.260: "the PP inversion can effectively reproduce results of the serial inversion" but only at a coarser resolution than the actual control vector, according to what is shown previously. This sentence is too optimistic as such.

AC: We have modified the sentence to address the concern of the reviewer:

"…shows that the PP inversion can effectively reproduce results of the serial inversion at regional scales"

- p.9 l.265-266: "a forward or adjoint TM5 CTM run of one year took about 15 minutes": it is a bit strange that the adjoint and forward codes take the same time to run - usually, the adjoint takes more time as it requires to recompute (or at least read) parts of the forward in sub-time-steps.

AC: The TM5 run used in this study is strictly linear. Therefore, the adjoint CTM run does not read or recompute parts of the forward CTM in sub-time-steps.

- p.9 l.270 seq.: the times for each step are indicated but nothing is said on the sink (see also comments on Section 2). Please add the information, even if it is only to state that it takes a very small computing time.

AC: We have added that the sink operater takes negligible time.

Section 4 Discussion

- p.10 l.307-309: "the need for a serial sequence of inversions to provide a time series of initial mole fractions imposes a limitation to the model resolution that can be used." This sentence is not very clear to me. Do you mean that the model's resolution must not be too fine so that the serial sequence of inversions does not take too long?

AC: We have modified the sentence:

“These numbers depend of course on the parallel efficiency of the model and the computing server. The need for a coarse resolution serial sequence of inversions to provide initial mole fractions fields limits the inversion period for which this method can be used.”

- p.10 l.310: "the wall time performance of the CAMS reanalysis inversions will improve in future". Taking into account the estimated gains in computing time given in Section 3, is it possible to give an order of magnitude of this expected improvement?

AC: We expect the wall time performance of a 30-year CAMS reanalysis inversion to improve by 5 to 10 times relative to the current CAMS inversion setup (which is not a serial inversion) if the PP implementation is similar to that used in this study: splitting the inversion period into annual block with 9-month overlaps. The actual improvement will depend on the number of sensitivity tests and checks performed and will be known after those CAMS inversion are performed.

- p.11 l.323: "optimizing emissions from large TRANSCOM regions". I understood from the description of the B matrix that the emissions were optimized at the resolution of the model's pixel (see also my comments on p.8 l.237-238). Here, it looks like the control vector contains the TRANSCOM regions. Please clarify.

AC: The inversion is performed at model pixel resolution. The emission estimates from the inversion are evaluated at TRANSCOM region scales. We have modified the sentence to address the confusion:

“We used a 9-month overlap in our test inversion setup. It was sufficient estimate the total emissions from TRANSCOM regions using the surface observations”

- p.11 l.334-336: "Furthermore, the performance gained by performing the inversions at higher resolution because of the improved computational performance will likely outweigh the accuracy loss due to the assumptions made in the PP method." This does not seem so likely to me. Do you have any references or examples of such a positive case?

AC: We do not have a reference for this. We assume that performing the inversion at a higher resolution will reduce the following errors:

1. Aggregation error: error in the distribution of emission at for example 1 x 1 degree grid that is not corrected by adjustment of the emissions state vector at 6 x 4 degree grid scale.

2. Model representation error: spatial and temporal smoothing intrinsic to the model resolution which prevents it from resolving finer-scale variability in the observations.

If PP method is implemented properly, with sufficient overlap and a sink operator S, the errors in PP should remain smaller than these errors.

- p.11-12 l.348-349: "These studies assume a quasi-linearity for the inversion as changes to the methane mole fractions are expected to remain small compared to the mean." This is not a definition of (quasi-)linearity. Please elaborate.

AC: We have added the following text to elaborate on this:

“The simultaneous optimization of OH with methane emissions introduces a non-linearity in the inversion because methane loss rate depends on the product of methane and OH mole fractions. However, the changes to the methane mole fractions are expected to remain small during the inversions. Hence, the non-linear effect is small and a quasi-linearity is assumed to solve the inversion analytically using the computation of the full Jacobian matrix of the CTM. Under a quasi-linearity assumption, OH can be optimized in a PP methane inversion by introducing annual OH scaling factors in the state vector and the methane lifetimes in the sink operator can be scaled in each iteration to reflect the corresponding OH adjustments.”

Section 5 Conclusions
- p.12 l.355: "An atmospheric inversion with a very large state vector is needed" This is not exactly true: many inversions are run with large regions and are able to use the available information on the tendencies, the North-South gradient, etc.

AC: We have modified the sentence:

“An atmospheric inversion with a very large state vector is needed optimize emissions using such long measurement records at a grid scale”

Figure 5
What is the link between the "2-sigma uncertainties of the prior emissions" and the description of B (p.7 l.195 seq.) i.e. 50% per grid cell per month plus the covariances (spatial and temporal correlations)?

AC: Yes, the $\pm 2\sigma$ prior uncertainties of the large regions shown in Figure 5 account for the spatial and temporal correlations (off-diagonal element of **B**) between the cells. We have modified text to clarify how **B** is constructed

“The prior covariance matrix **B** is constructed as follows. The diagonal elements of **B** are constructed assuming $\pm 1\sigma$ uncertainties of 50 % of the emissions per grid cell per month. The off-diagonal elements are constructed by assuming the emissions to be correlated temporally using an exponential correlation function with an e-folding time scale of 3 months, and spatially with a Gaussian correlation function using a length scale of 500 km (Houweling et al., 2014).”

Technical corrections
- in the introduction (and maybe other places also): "CTM's" (saxon genitive, I guess) must be changed to "CTMs" (plural of CTM)
- p.2 l.63: "emissions adjustments" -> emission adjustments?
- p.4 l.122: "scaler" -> scalar?
- p.4 l.124: "we use a CTM block sensitivity vector h_k distribute global emission changes more precisely" -> we use a CTM block sensitivity vector h_k TO distribute THE global emission changes more precisely?
- p.7 l.200: "The emissions in 2008 applied to every year in the inversion period" -> The emissions of the year 2008 are used for every year of the prior?
- p.7 l.208: ". sufficient for our test inversion (Section 3)" -> does this part of sentence goes with the previous one? If so, change "." to ",". Also, why is there a reference to Section 3 inside Section 3? Please check.
- p.7 l.224: "as good as" -> as well as?

AC: We have implemented the technical corrections suggested by the reviewer.

References

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