Comment on gmd-2021-339
Anonymous Referee #3

The authors have developed a kind of “window-splitting” scheme for a variational inverse analysis of atmospheric CH4, which can be performed by parallel computing. For a multi-decadal analysis of long-lived species such as CO2 and CH4, a variational inverse analysis would be time consuming even when a massive amount of computational resources are available. This is because a variational analysis is basically a serial computation algorithm, which requires iterative calculations. In this regard, the developed method is worthy of publication from GMD, though its basic idea is already published by Chevallier (2013). Before publication, however, the reviewer would like the authors to revise the manuscript considering comments described below.

It is difficult to follow the description of the scheme, whose major reason is that many matrices and vectors are not written in bold fonts. This is very confusing. Furthermore, the reviewer strongly recommend that the author should clearly describe what is new and different from the original scheme of Chevallier (2013).

Although the reviewer is not a native English speaker, the reviewer thinks that the English writing of the manuscript has much room to improve. Therefore, a native check is also recommended.

The authors claim that the developed scheme is effective for a long-term inverse analysis in terms of wall clock time. The reviewer has no doubt about it, but would like the authors to discuss its relative effectiveness comparing with other approaches. For instance, a MPI parallelization (much more scalable parallelization than OpenMP) on the transport model could also shorten the wall clock time.

Specific comments:
L11: “variational (4DVAR)” => “four-dimensional variational (4DVAR)”
“by a factor of 5” its computational effectiveness should be also described. How much computational resources are increased?

“CAMS (Copernicus Atmosphere Monitoring Service)” => “Copernicus Atmosphere Monitoring Service (CAMS)”

“CTM (chemical transport model)” => “a chemical transport model (CTM)”

Which is “this study”, the study by the authors or the one by Saunois et al.? Maybe it is the latter, but it should be clarified for more general readers.

“representing the sensitivities by a statistical ensemble” is not clear.

“is obtained” would be better than “is computed”

“computational efficiency” might be inappropriate, because the computational resources used in the inversion were increased.

Chevallier (2013) named the scheme as “physical parallelization (PP)”, but the authors here named their scheme as “physical parallelization for variational inversion (PPVI)”. Are they the same? If that is the case, it would be better to use PP rather than PPVI to respect the original idea of Chevallier (2013).

Somewhere in Introduction: More introduction about CH4 inverse analyses other than Saunois et al. (2020) would be beneficial.

transpose “T” is missing. “(x-x_a)B^{-1}(x-x_a)” => “(x-x_a)^T B^{-1} (x-x_a)”

“(H(x) - y)R^{-1}(H(x) - y)” => “(H(x) - y)^T R^{-1} (H(x) - y)”

In here” => “Here”

the a” => “the”

Why can the CTM that calculates the initial mole fraction fields be performed at the coarser resolution?

What is the “methane perturbation”?

Please describe how the mole fraction conversion factor (=0.361) is derived.

Eqs. (3)-(5): Are \(c_0\), \(x_i\), \(n_i\) scalars or vectors? If they are scalars, are they the global totals?

Please elaborate the sufficiency of the e-folding decay function, because this might be the new and different from the original scheme of Chevallier (2013).

What is “the adjoint test”? Please elaborate it.

“uniform” is better than “unity”, isn’t it?

Are “78 ppb” and “28 ppb” the results of serial or PPVI?

For both inversions, the good fit .... a gradient reduction of 1000 is sufficient” The fit to the observations cannot be used to determine the sufficiency of the convergence.
L207-208: “The parallelized ... in the serial inversion” is not clear.

Section 3.1: One may want to see differences of more small scales (e.g., flux patterns, seasonal cycles).

Section 4.1: This section would be better to be moved to Introduction.

L257: “if future” => “in future”?

L278: Please spell out “SWIR” and TIR, because they appear first here.

L280-281: “These studies ... small in an inversion.” Is not clear.

L282: Does “the methane lifetimes in the S operator would be scaled in each iteration” mean that S is included in the control variables?