General Comments

Bisht et al present a data assimilation system for local ensemble transform Kalman filter, and evaluate that through OSSEs, particularly testing three covariance inflation methods (fixed multiplicative, relaxation to prior spread, and adaptive multiplicative) and two observing networks (surface dense network and GOSAT satellite network). This manuscript describes several interesting findings. I have three concerns.

1. This OSSE does not account model transport error, which would result in over-optimized solutions.
2. The number of ensemble members is not sufficiently greater than the dimension of the state vectors, which might bias the inversion performance interpretation.
3. Several sections require clarifications, as in the following "Specific Comments".

Thank you for reviewing the manuscript and providing us useful comments and suggestions.

Our reply to Point number 1

We mentioned transport error limitation in our OSSEs at the end of Section 5 of our revised manuscript as follows:
"We have not accounted for the transport error due to meteorological fields in this work (Patra et al. 2011), in case of real observations data assimilation a week-long window may introduce transport errors in CH4 analysis because of nonlinear growth of ensemble perturbations."

Patra, P. K., Houweling, S., Krol, M., Bousquet, P., Belikov, D., Bergmann, D., Bian, H., Cameron-Smith, P., Chipperfield, M. P., Corbin, K., Fortems-Cheiney, A., Fraser, A., Gloor, E., Hess, P., Ito, A., Kawa, S. R., Law, R. M., Loh, Z., Maksyutov, S., Meng, L., Palmer, P. I., Prinn, R. G., Rigby, M., Saito, R., and Wilson, C.: TransCom model simulations of CH4 and related species: Linking transport, surface flux and chemical loss with CH4 variability in the troposphere and lower stratosphere, Atmos. Chem. Phys., 11, 12813–12837, https://doi.org/10.5194/acp-11-12813-2011, 2011b.

Our reply to Point number 2

In our LETKF data assimilation system the localization approach is used to mitigate spurious correlation due to much smaller ensemble size than the degrees of freedom of the system. In our revised manuscript we added the discussion on localization approach (Section 3.3, Paragraph 2):
"In this study, the CH4 observations are assimilated by applying the observation error covariance localization (Kotsuki et al., 2020) to reduce the spurious spatial correlation due to smaller ensemble size than the degrees of freedom of the system……"

Our reply to specific comments (point number 3): for the specific comments our replies are given in black fonts for your comments in red.

Specific Comments

L43: "Cl in the stratosphere". Suggest including Cl in the troposphere.

Reply: Thank you for the suggestions.

CH4 loss to Cl takes place in the marine boundary layer (MBL), where sea salt is abundant, but CH4 is also destroyed in the stratosphere by reaction with Cl (e.g. Röckmann et al., 2004; McCarthy, 2003). In the modified sentence we didn't specify troposphere or stratosphere because our model simulations consisted of these in both the layers but we have not explicitly included Cl from sea-salt sources. We rewrite it as follows:

"other loss processes include oxidation by soil, and reactions with O1D and Cl"

Röckmann, T., J. ‐ U. Grooß, and R. Müller (2004), The impact of anthro- pogenic chlorine emissions, stratospheric ozone change and chemical feedbacks on stratospheric water, Atmos. Chem. Phys., 4, 693–699.

McCarthy, M. C., Boering, K. A., Rice, A. L., Tyler, S. C., Connell, P., and Atlas, E.: Carbon and hydrogen isotopic compositions of stratospheric methane: 2. Two-dimensional model results and implications for kinetic isotope effects, J. Geophys. Res., 108, doi:10.1029/2002JD003183, 2003.

L122: Typo, "the ensemble forecast of CH4 concentrations"

Reply: Corrected.

L79: "Advanced". Could you please specify what is the advanced aspect of this study, comparing to the previous studies using the same model? Is it the setup of the multi-window optimizing framework, or these inflation methods, or others?

Reply: The advanced aspect of this study includes the use of different inflation methods in our research and the simultaneous estimation of atmospheric concentration and surface fluxes of CH_4.

L188: "by 30%". Unclear if this is uniform bias. According to the later text, the perturbation is not uniform. Could you please specify the way to combine this "30%" with the following regional/grid level perturbation?

Reply: The systematic bias of a prior flux against true flux is assumed to be 30%. Besides, random perturbations equivalent to standard deviation of 6-8 % are added to the a priori flux as the initial ensemble spread. We modify the sentence such as:

"An initial perturbation with standard deviation of approximately 6-8% is applied to the a priori flux as the initial ensemble spread."

L196: "Experiment1". The word is misleading. Confused the readers if these experiments are corresponding to the experiments in section 4.1 and 4.2 (in fact, they are not).

Reply: We attempt to simplify it by modifying the sentences as follows (L202-207):

"This study performs two LETKF data assimilation experiments. In these experiments, we provided initial perturbation on regional basis over land (53 different land regions; Chandra et al., 2021) and at every grid over ocean, no spatial error correlation between grid points is considered among ensemble members. However, in Section 4.2.5, we also discussed the sensitivity of CH4 data assimilation by providing initial ensemble spread at every grid by considering horizontal spatial error correlation between grid points among ensemble members, with a global mean correlation of 20%."

L196: "regional basis over land" and "every grid over ocean". Please explain why emissions over land and over ocean are perturbed differently.

Reply: We demonstrated in our CH4 LETKF sensitivity to initial ensemble spread experiment (Section 4.2.5) that, the estimated error between analysis and true fluxes with grid-based initial ensemble spread (both over land and ocean) is significantly larger (25%) than region-wise (region-wise over land and grid-wise over ocean) ensemble spread.

L207: "Only surface layer CH4 concentrations are used". Both over land and ocean? Please explain if the "dense observation network" include all surface grids or a collection of surface networks. If it is the first case, the word "dense observation network" is confusing.

Reply: It include all surface grids. We replace "dense observation network" to "dense observation data" in our revised manuscript.

L208: "added a constant measurement uncertainty of 5ppb". Please explain the way to add this 5 ppb (uniformly increase/decrease 5 ppb?). Also, typo, space between "5" and "ppb".

Reply: Uniformly increase 5 ppb uncertainty is being added. Typo corrected.

L236: "3.4 Experiment2". In experiment 1, "dense observation formulation", the author added measurement uncertainty of 5 ppb. Please explain why experiment 2 has no observation error, given the fact that satellite observations have larger uncertainties than measurements of surface sites.

Reply: It is already mentioned in the manuscript (L260-262) as follows: "we added the same retrieval (XCH4) error as GOSAT to the XCH4 (ACTM simulated) to make the OSSE more realistic and then attempt to estimate the true fluxes."

L406: "Machine learning tools could be used to". Machine learning comes from nowhere. Please explain why it would help.

Reply: We remove this statement because machine learning is beyond the scope of this paper.