Reviewer #2

Comment [2-1]: General comments: The authors extend their previous coarse-grid global inversions to a fine-grid regional scale. They optimize methane emissions and 2010-2017 emission trends in North America by in situ (GLOBALVIEWplus CH_4 ObsPack) and satellite (GOSAT) observations, through analytical inversions using log-normal error forms. They point out large emission underestimates in the oil sector by a factor of 2, and a peak of CONUS anthropogenic emissions in 2014. The paper is well written. The methods are clearly described, and the results are well discussed. I support publication, but with a major concern and some minor suggestions.

Response [2-1]: We thank the reviewer for the positive and valuable comments. All of them have been implemented in the revised manuscript. Please see our itemized responses below.

Comment [2-2]: My major concern is that this study lacks independent evaluation. The authors compare the posterior simulation and prior simulation against the observations used for the inversions, and the improvements against GOSAT are weak. I am curious about the evaluation against the independent dataset, such as TCCON or other local in-situ measurements.

Response [2-2]: Thank you for pointing it out. We have added independent evaluation with the five TCCON sites in US. We now state in the text: "Independent evaluation with the ground-based column observations from the Total Carbon Column Observing Network (TCCON) (Wunch et al., 2011) further shows that the mean model bias at five sites in CONUS decreases from 5.2-14.0 ppbv in the prior simulation to 1.0-13.5 ppbv in the posterior simulation."

Reference

Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V., and Wennberg, P. O.: The Total Carbon Column Observing Network, Philos. T. R. Soc. A, 369, 2087–2112, https://doi.org/10.1098/rsta.2010.0240, 2011.

Comment [2-3]: Specific comments and technical corrections. Row 199: What are the treatments for the initial conditions of the global simulations?

Response [2-3]: We now state in the text: "The initial methane concentration fields on 1 January 2010 are from Lu et al. (2021) which have been adjusted to have an unbiased zonal mean relative to GOSAT observations, such that model discrepancies with observations over our 2010-2017 simulation period can be attributed to model errors in emissions instead of errors in initial conditions."

Reference

Lu, X., Jacob, D. J., Zhang, Y., Maasakkers, J. D., Sulprizio, M. P., Shen, L., Qu, Z., Scarpelli, T. R., Nesser, H., Yantosca, R. M., Sheng, J., Andrews, A., Parker, R. J., Boesch, H., Bloom, A. A., and Ma, S.: Global methane budget and trend, 2010–2017: complementarity of inverse analyses using in situ (GLOBALVIEWplus CH_4 ObsPack) and satellite (GOSAT) observations, Atmos. Chem. Phys., 21, 4637-4657, http://doi.org/10.5194/acp-21-4637-2021, 2021.

Comment [2-4]: Row 254: Are the Jacobian matrix for the boundary conditions constructed in the same way as the grid-level emissions?

Response [2-4]: Yes. We have clarified in the text: "We construct K by conducting GEOS-Chem simulations where each element of the state vector (methane emission and model boundary correction) is perturbed separately."

Comment [2-5]: Row 297: The error standard deviations for boundary conditions are 10 ppb in the base inversion and 5 ppb in the sensitivity inversions. These are much smaller than the error standard deviations for emissions. How sensitive are the results if applying a larger error standard deviation for boundary conditions?

Response [2-5]: We find that compared to the default 10 ppb used in the base inversion, using 5 ppb as the error standard deviations for boundary conditions leads to less than 1% difference in the resulting methane emissions in US, Canada, and Mexico, indicating its small sensitivity to the main conclusion. We did not use a larger error standard deviation because we have much more confidence in the boundary conditions than emissions, as it is generated from the global simulation using methane emissions and sinks optimized by GOSAT observations in our previous work (Lu et al., 2021). These global methane fields are unbiased to global zonal mean observations for 2010-2017, with some residual bias for individual years but even those are less 5 ppbv for the northern mid-latitudes (Fig.6 in Lu et al., 2021).

We now state in the revised text "Lu et al. (2021) show that their optimized simulation is unbiased in comparison to global zonal mean observations for 2010-2017 but we still find some residual biases for individual years up to 5 ppbv.".

Comment [2-6]: Row 317: The observation error standard deviations for in-situ data are ~2× of that for GOSAT, and the total number of observations for in-situ data is 0.4× of that for GOSAT. Readers may be curious about the results if applying two regulation parameters separately to in-situ and GOSAT data.

Response [2-6]: Yes, we indeed determine and apply two regulation parameters separately to the in-situ and GOSAT data. We now clarify in the text: "Here we determine the regularization factor γ separately for in-situ and GOSAT data following Lu et al. (2021), and find that γ = 1 is best for the both. We also conduct a sensitivity inversion using γ = 0.5 for the GOSAT observation terms (while keeping γ = 1 for in-situ data terms in the joint inversion) as adopted in Maasakkers et al. (2021)."

Comment [2-7]: Row 417: "This may reflect the underestimation of CO_2 over the Los Angeles Basin". Typo, CH_4 rather than CO_2?

Response [2-7]: This is indeed the model CO_2 used in the proxy GOSAT retrieval, rather than CH_4. We now clarify in the text: "This may reflect the coarse resolution of model CO_2 used in the proxy GOSAT retrieval that leads to underestimation of CO_2 (and hence methane) over the Los Angeles Basin (Turner et al., 2015; Maasakkers et al., 2021), and/or complex topography."

Reference:

Turner, A. J., Jacob, D. J., Wecht, K. J., Maasakkers, J. D., Lundgren, E., Andrews, A. E., Biraud, S. C., Boesch, H., Bowman, K. W., Deutscher, N. M., Dubey, M. K., Griffith, D. W. T., Hase, F., Kuze, A., Notholt, J., Ohyama, H., Parker, R., Payne, V. H., Sussmann, R., Sweeney, C.,

Velazco, V. A., Warneke, T., Wennberg, P. O., and Wunch, D.: Estimating global and North American methane emissions with high spatial resolution using GOSAT satellite data, Atmos. Chem. Phys., 15, 7049-7069, http://doi.org/10.5194/acp-15-7049-2015, 2015.

Maasakkers, J. D., Jacob, D. J., Sulprizio, M. P., Scarpelli, T. R., Nesser, H., Sheng, J., Zhang, Y., Lu, X., Bloom, A. A., Bowman, K. W., Worden, J. R., and Parker, R. J.: 2010–2015 North American methane emissions, sectoral contributions, and trends: a high-resolution inversion of GOSAT observations of atmospheric methane, Atmos. Chem. Phys., 21, 4339-4356, http://doi.org/10.5194/acp-21-4339-2021, 2021.

Comment [2-8]: Row 425: “For GOSAT the improvement is less apparent from the comparison statistics, because the prior simulation already has a low mean bias MB = -0.5 ppb, and the prior RMSE is only 6.9 ppb (which decreases to 6.5 ppb). However, we see from Figure 5 a significant whitening of the noise with reduction of regional-scale biases.” This sounds like to contradict itself. It first presents that the regional mean bias of the posterior simulations is 0.6 ppb, larger than the -0.5 ppb in the prior simulations, then indicates that the posterior simulations' regional-scale bias is less than prior simulations in the map.

Response [2-8]: Thanks for pointing it out. We have revised the text accordingly: “For GOSAT the improvement is less apparent from the continental-scale comparison statistics, because the prior simulation already has a low mean bias MB = -0.5 ppb and a small RMSE of 6.9 ppb. However, we see from Figure 5 that the small mean bias reflects an offset between high bias in western US and Canada and low bias in the central and eastern US. The inversion results in spatial whitening of this bias.” We have also added independent evaluation against TCCON sites (Response [2-2]).