The author uses the GOSAT satellite-based columnar and surface observation of CH_4 to optimize CH4 emissions for 2010-2018, focusing on South America, especially the Amazon river basin and Brazil. The author reported an increasing emission trend for optimizing emissions over Brazil. The author further observed the strong trend during the wet season and attributed the wetland emissions as the main driver. The author used the bottom-up model to investigate the causes of variations in wetland CH4 emissions. However, the fluxes neither match with variation in annual fluxes nor the positive trend in the inversion emissions. The author reported no change in fossil fuel emissions for the same period. Overall, the results are impressive, with detailed and careful investigations. The paper would greatly benefit from a full revision to ensure the authors' key messages are easier to understand. Below I outline my substantive and minor comments.

We thank the reviewer for his/her comments, which have significantly helped to improve the paper and clarify our results and message. We hope that we have addressed these concerns appropriately. Our point-by-point response is given below, highlighted as blue text.

Substantive Comments:

1) The paper looks lengthy, which make it difficult to understand key messages clearly. I would suggest revising the structure and moving the proxy information (e.g., Detailed of Bottom-up simulations) in the supplement part, as indicated by Luke Western.

We have generally attempted to shorten the main text where possible and have also moved some sections to the supplement as suggested here and by other reviewers.

2) Estimated error covariance is as essential as estimated fluxes, yet these are never shown. I would like to know what the decrease in error covariance is between the prior and posterior.

OK, we have included a plot of this within the Supplementary Material.

3) The accuracy of model transport plays a crucial role in interhemispheric gradients in CH4 near the Earth's surface and vertical gradient in the troposphere and stratosphere since the total column consists of 40% of stratospheric air, depending on seasons and latitude. Thus, it is crucial to test the accuracy of the troposphere and stratosphere vertical transport before optimization. Too fast transport of too slow transport in the model will directly hinder the optimization results. The vertical gradient in the stratosphere becomes more critical for simulating XCH4 data. Along with transport, the validation of chemical loss is also vital for optimizations. It would be useful if the authors reported the methyl chloroform e-folding lifetime as a way of assessing the prior OH and another proxy (SF6) simulations for validating the inter-hemispheric and vertical transport. Even if this is addressed in an earlier paper, some Supplementary or acknowledgement plots would be useful.

You are correct that model transport errors will likely play an important role in the results obtained in this work. We attempt to account for a significant proportion of model transport error, particularly in the vertical distribution, through addition of a model – satellite bias term derived from a previous inversion based on surface flask observations of CH_4, whilst acknowledging that there will very likely remain some model error that is unaccounted for. We have previously investigated the

simulated interhemispheric gradient (IHG) in TOMCAT ourselves through SF_6 simulations (e.g. Wilson et al. (2014)), and have also taken part in many multi-model intercomparisons, including for CH_4 (e.g. Patra et al. (2011), Thompson et al. (2014)). In all cases the model performs well, particularly in comparison with other similar models. In Patra et al. (2011), the TOMCAT model used the OH fields that we use in this study (TOMCAT CH_4 lifetime: 9.98 years), and also applied them to simulations of MCF, matching the observed MCF lifetime well (TOMCAT: 4.71 years compared to observed value of 4.9± 0.3 yr (Prinn et al., 2005)). However, it is true that the IHG in TOMCAT tends to be slightly overestimated compared to observations, and this will likely affect our results. We have added this caveat into the main text in the model description section and referred the reader to the references discussed here. We feel, however, that replications of figures from previous studies within this manuscript would only act to unnecessarily increase the length and complexity of this paper and hope that referring the reader to previous work is a satisfactory response.

4) It is difficult to follow how authors get the uncertainty ranges (e.g., Fig. 4a, b). It would be generous to the reader if you state clearly this information in the text.

This information is included in Section 2.2.1 and has been clarified.

5) It is currently difficult to understand the spatial distribution of sectoral emissions to understand their role in the study region's methane emission changes. The spatial distribution of prior emissions (probably Figure in the Supplementary Section), mostly like wetland emissions, Enteric fermentation and manure management emissions, biomass burning, and Fossil Fuel, will help the reader.

OK, we have included a plot of this within the Supplementary Material.

6) It would be not easy to pinpoint the role of wetland emission in increasing trend compared to the decreasing wetland areas for the contemporary period. In such a case, the role of bottom-up simulations is challenging to connect the dots. What about the role of Enteric fermentation and manure management, and agricultural emission, which are also increasing over Brazil as per the updated version of EDGAR inventory (EDGARv4.3 and latest version)? Investigating such dimension will make the study interesting. The spatial sectorial emission map of or trend in the prior sectorial emissions can help excavate such information.

Yes, as suggested by this reviewer and others, we have changed the tone of the main text to make it clear that wetlands are not the only potential culprit for changes in the total flux and that using our methodology does not allow us to further speculate on the source of the variations. We have included the reference to the latest EDGAR dataset, and have also included the possibility that biomass burning sources could be changing outside of the dry season. The trend in the prior emissions used in our study is negligible, so we decided not to include it as a figure.

7) Using the histogram for the validation does not give enough information about how well the optimized emissions improved the fitting of the observed trend? It would be useful for the reader to show the observed and fitted XCH4 time-series (say over the region shown in Fig. 5) and the time series over two different altitude range of aircraft as shown in the Figure. 6.

We have included a plot of observed (GOSAT) and simulated XCH4 over the Amazon basin and Brazil in the Supplementary material (Figure A4). The aircraft observations will be published in a separate upcoming manuscript.

Specific comments.

L32: "Cannot match. . ." –> "Neither match. . .". Since you are using "nor" conjunction

Done.

L31: "Much of CH4. . .." Consider giving the numerical fraction.

If this refers to line 39, we have adapted the text as follows:
"Approximately 90% of the CH_4 that is emitted into the atmosphere is eventually destroyed through reaction with the hydroxyl (OH) radical and the remainder is lost to other smaller sinks,…"

L43: "remove "to the atmosphere" and "in the atmosphere". Can state like "However, the magnitude of global sources and sinks are still not well quantified..."

Done.

L50: The period is not matching with the previous studies.

Yes, we have changed '2007' to '2006'.

L64: The authors could also cite Chandra et al. JMSJ. (2021) (https://www.jstage.jst.go.jp/article/jmsj/advpub/0/advpub_2021-015/_article), who also found an increase in both biogenic and fossil fuel emissions. Naus et al (2020) (https://doi.org/10.5194/acp-2020-624) and Patra et al. (2021) (https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2020JD033862) also studied the OH trend using CH3CCl3 observations and observed no significant trend.

Thank you for highlighting these important recent references, we have included them in the text.

L58: Point 1st and 3rd have more or less the same conclusion. Consider it to merge. So basically, you will have two points of source uncertainty and sink uncertainty.

We have merged and slightly rewritten the two points.

L64: Better to state more conclusively. Worden et al. (2017) suggest that the equal contribution of both FF and biogenic sources is also possible if pyrogenic emissions are decreased for the same period.
We have added this information.

Section 2.1.3. It is not mentioned in this Section the use of aircraft measurements in this study. After reading this paragraph, I thought the aircraft measurements are also used in optimization. The information regarding the use of these observations for the validation purpose is coming later.

Yes, we have added text at the start of this section to clarify this.

L215. Not clear 250% of what? Is uncertainty 250% of prior emissions?

Yes, we have now included this information.

Section2.2.2 Consider showing all the sectoral emissions time series for the study region (maybe in Supplementary?)

We have included maps of the sectorial emissions. There is actually very little variation in time in the prior emissions, so we have decided not to include a figure showing the sectorial timeseries.

L304. "The posterior residuals show no significant trend or seasonality" -! Over where? Over Amazon Basin or the whole of South America?

We have clarified that residuals show no trend within either region.

L305. Similar emission maps for each sector in Supplementary will help the reader to understand the dominant emission sources over different regions.

We have included maps of the sectorial emissions in the Supplementary section.

L335. Figure S1 is not shown. Maybe you are talking about Figure A1. Correct others also throughout the manuscript.

Done.

L345. How did you calculate the uncertainty?

This is described in Section 2.2.1

L346. "This means. . ..." How does the previous sentence follow this conclusion? at least gives the number. . .. Consider reformulating

The sentence actually says ‘this *mean* flux’ but we accept that this could be unclear. We have changed it to ‘Our mean flux value’.

L353. What is the shoulder season?

This ambiguous phrase has been removed.