We thank both reviewers for providing useful comments to the submitted manuscript. We also thank Janne Hakkarainen for spotting a typo in Appendix B that we have now fixed. Below we detail our responses to individual comments and, if relevant, how we altered the revised manuscript.

Response to reviewer 1

Our initial response was in late June 2022. Here we include more detailed responses

Major comments

A crucial aspect of this work that is missing is the evaluation. There seems to be no evaluation of the results using independent observations or techniques like k-fold cross validation. The work addresses an important and timely topic but, in this reviewer's opinion, the main claims in the manuscript (that the growth is driven by emissions, not chemistry) do not seem supported by their numerical experiments.

We have indirectly evaluated our results for 2020 by inferring methane emissions using the NOAA surface data, which provide consistent results on global and continental spatial scales. We will make this point clearer in the revised manuscript. When we first responded to this comment in June 2022, we did not have access to NOAA data for all sites in 2021.

We have now evaluated our posterior emissions using independent TCCON XCH4 measurements (v GGG2020) for 2020 (Figure C1) and the first half of 2021 (not shown). We find that the annual mean model minus TCCON XCH4 values for 2020 are within 10 ppb for 16 (out of 18) sites with standard deviation between 5 and 18 ppb but typically < 10ppb. This is consistent with our evaluation of data from 2009-2019 (Feng et al, 2022), building on previous studies that have included substantial evaluation using a range of in situ and remote sensing data. Our preliminary analysis of an incomplete year of TCCON data in 2021 shows similar model performance. To address this point, we have added Appendix C that includes a figure showing the mean comparison statistics.

The suggestion to use k-fold cross validation is interesting. We have used this method in other less computationally intensive applications but to our knowledge it has not been used to evaluate a global inversion. We are unconvinced this will add much to our narrative, especially given the focus of the review is on 2020.

***The major concern this reviewer has with the manuscript is that the title and central claims don't seem supported by their data. The main scientific claim (and their final conclusion) is that the record-breaking methane growth rates in 2020 and 2021 were driven by emissions, not chemistry. This claim certainly seems plausible (if not likely), but their experiments do not seem suficient to justify that claim. In this reviewer's opinion, the ideal way to conclude as to the relative importance of emissions and chemistry would be to include both emissions and OH in the state vector for their EnKF. That would result provide a straight forward assessment of the relative role of each process*.**

The sensitivity experiment we used to quantify the impact of OH is admittedly a brute-force approach but in the absence of rigorous constraints on OH concentrations we felt this was a transparent approach that is easy to understand. As part of our revision, we performed new experiments that assume that the OH reduction in 2020 roughly followed the observed temporal and spatial changes for tropospheric ozone (Ziemke et al., 2022). Again, we find that increased emissions largely explain the large global mean growth of atmospheric methane in 2020. This is described in Appendix D.

An alternative way would indeed be to include both emissions of methane and OH in the state vector, and this is what we have done in the revised manuscript. These additional calculations are described in Appendices D and E and summarized in the main text.

We have conducted new experiments by including monthly scaling factors for OH concentrations over 18 global regions as part of state vector (Appendix E). The resulting emission increase between 2019 and 2020, is now about 25% smaller than our control run that solves for methane emission estimates using OH climatology. This result is consistent with emissions being primarily responsible for the anomalous global atmospheric methane growth rate in 2020. As the reviewer will be aware there are also still gross assumptions associated with this OH-methane emission inversion and the resulting posterior results do not provide a "straightforward assessment" of OH changes due to the limited information content of the satellite and in situ atmospheric methane data. Methane inversions that also estimate OH, typically quantify OH on large spatial scales and therefore will be unable to identify localized changes in OH that we anticipate happened during the Covid-related lockdowns.

The ideal approach would be to include the methane emissions in a state vector as part of a full-chemistry data assimilation system that also consider more appropriate constraints on OH. But of course, this problem becomes progressively more complex (non-linear) and more intractable (associated with more chemistry constituents and more data).

See also our response below, which discusses larger OH perturbations.

The argument presented in this manuscript, as this reviewer interpreted it, is as follows:

the authors conducted a global inversion at 2◦ × 2.5◦ resolution with an EnKF from 2019-2022. This inversion assumes constant OH fields for the 3-year window. The authors find changes in the magnitude and spatial patterns of methane emissions. the authors compared these emission changes to rainfall, GRACE groundwater, and temperature. The largest correlations were 0.5-0.6 (representing 25–35% of the variability). the authors conducted a second global inversion with the same setup but reduced OH by 5% through 2020, where the largest COVID changes occurred.

We are certainly more confident in the geographical distribution of emissions rather than their attribution. A more detailed study about the attribution of those emissions will be forthcoming but it is outside the scope of this current study.

Saying that, the spatial and seasonal variations in tropical methane emissions are consistent with those reported for earlier years (e.g., Lunt et al, 2019, 2021; Feng et al 2022; Wilson et al, 2021) that showed the response of methane emissions was consistent with microbial sources. However, we think imperfect attribution does not detract from our key message: large emissions are predominantly responsible for anomalous atmospheric growth rates in 2020 and 2021.

The authors show the difference in emissions resulting from these cases but it is not clear to this reviewer which result is better. The differences seem to be central to their conclusions as indicated in the last two lines of their abstract ("Based on a sensitivity study for which we assume a conservative 5% decrease in hydroxyl concentrations in 2020...we find that the global increase in our a posteriori emissions in 2020 is ~22%

lower than our control calculation. We conclude therefore that most of the observed increase in atmospheric methane during 2020 and 2021 is due to increased emissions.") but I could not discern how they concluded why one was better than the other. Specifically, it is unclear why the control calculation is the correct answer here.

We did not conclude one was better than the other. We concluded that a 5% drop in OH was too large using our perturbation approach, which described only 22% of the emission from our control run. Consequently, most of the atmospheric growth in 2020 and 2021 was due to emissions.

Our additional sensitivity calculations, including an OH inversion, are all consistent with our original hypothesis about increasing emissions in 2020 being the most likely culprit for the unprecedented global atmospheric methane growth rate. We will also put this discussion in the context of higher, localized reductions in OH as raised as a discussion point by this reviewer (see below).

Evaluation and/or overfitting

Two common methods for evaluating the performance of optimization schemes are to: 1) evaluate against independent observations or 2) perform k-fold cross validation. Neither of these were included here. This is something that should be included for all their cases with an inversion analysis to ensure that one is not overfitting for a particular inversion

This study is a two-year extension of recent work (Feng et al, 2022), including substantial evaluation of the results that build on previous studies. In an earlier response (see above) we have described our evaluation with TCCON data for 2020. We have also indirectly evaluated our 2020 results using an inversion constrained by NOAA data that leads to consistent results.

Since we use an ensemble Kalman filter the error characterization is a natural extension of our analysis. Using the cost function weights, we find no evidence to suggest we have overfitted the data. We have added text to that effect in the revised manuscript.

OH is inconsistent with other work

The authors chose a 5% reduction in OH based on Laughner et al. (2021). However, Laughner et al. (2021) was a review/synthesis paper that took global mean OH changes from Miyazaki et al. (2021; doi:10.1126/sciadv.abf7460) and used them in a box model. This reviewer wonders how large the global mean OH changes are in this manuscript from Feng et al.? My suspicion is that they are quite a bit smaller than what was reported in Miyazaki et al. and Laughner et al Additionally, the OH chemistry is highly non-linear and Miyazaki et al. discuss how OH and ozone actually increase in some regions despite the NOx reductions. Using OH fields from Miyazaki et al. would be a much better way of testing if the OH simulated in that work impacted the methane burden. Essentially, this reviewer does not think the OH sensitivity run designed here accurately portrays the OH changes that others have found. Data supporting the choice of OH runs used here would help assuage these concerns.

Our sensitivity experiments are designed to examine how large-scale reductions in OH (associated with Covid-19 lockdowns) affects our methane emission estimates inferred from atmospheric methane measurements.

First, it was remiss of us not to include Miyazaki et al, 2021 for which we apologize. This primary reference was added to the revised manuscript. Figure S8A from this paper shows localized OH reductions of 15-20% for May 2020, although the authors note that this reduction can be as large as 30% on a grid basis. This represents one of the early months of the shutdown when we expect the largest reduction in emissions, so we expect global mean annual changes to be smaller. We also note that Miyazaki et al, 2021 (as with others) do not consider the coincident changes in non-methane hydrocarbons (NMHCs) that will also affect non-linear ozone chemistry, including the production and loss of OH. Large coincident reductions in formaldehyde, for instance, a proxy of NMHCs, have since been reported in other studies (e.g., Sun et al, 2021).

We did a number of simulations in preparation for this manuscript. We used 1% and 5% global reductions in OH but penalized regions that would not have been impacted by reductions in nitrogen oxide emissions from Covid. We instead chose to reduce OH only over regions where there are substantial anthropogenic CO_2 emissions, resulting in a distribution similar to Miyazaki et al, 2021.

In response to this reviewer comment, we prepared a calculation in which we decrease OH by an additional 25% of eastern China for March 2020 (the peak of emission reductions). We find that this month-long perturbation can explain an additional 0.26 Tg of methane emissions for 2020, providing some indication of how a larger, localized reduction in OH will impact global changes in methane emission estimates. In the revised manuscript, we have reported the result of an additional calculation (Appendix D) that adopts an OH reduction pattern following changes in tropospheric ozone (Ziemke et al, 2022). We find the result is similar to the results from our other sensitivity test – the emissions needed to explain the atmospheric growth of methane in 2020 is about 25% lower than our control when we consider a drop in OH.

We have added a broader discussion of this kind of OH perturbation (Appendix D) and now included an OH inversion (Appendix E) in the revised manuscript. However, we hasten to add that this reviewer comment should be tempered by the uncertainties associated with previous studies that do not accurately describe the photochemical perturbation associated with the Covid-19 shutdowns. We added text that describes caveats associated with both approaches.

GOSAT proxy observations

The authors use GOSAT proxy observations. This means that the methane concentrations will be dependent on the CO2 concentrations. However, it seems like the authors use CO2 simulations with monthly emissions through 2019. Therefore the CO2 could lead to a bias in their methane concentrations during COVID due to the reduction in CO2 emissions. This would be most pronounced in urban areas.

As described in Parker et al., 2020, the GOSAT data use an ensemble of CO_2 models based on atmospheric in situ inversions. For recent years, when updated model inversions based on in situ observations are not typically readily available on the timescales required for data processing, we have used CO_2 values from previous years that have been incremented by the NOAA global growth rate. To ensure that this does not introduce an error due to model CO_2 values, an inversion is performed that directly uses the XCH4/XCO2 ratio, without relying (explicitly or implicitly) on the model CO2. We have clarified this point in the revised manuscript.

3.7. This reviewer was also very confused by the description of the data used in places. For example, when describing a sensitivity study the authors mention using proxy GOSAT XCH4 data (Line 90) but the main inversions also seem to use proxy GOSAT data.

We use two approaches: 1) the proxy ratio (XCH4/XCO2) directly without need to assume a prior model for XCO_2 to multiply out to obtain XCH4, which is described in Feng et al, 2022; and 2) the proxy methane XCH4. In addition, we have also assimilated in situ methane data in both approaches. As a result, we find both approaches lead to generally consistent results. We will amend the text to make this point clearer.

<u>Minor Comments:</u>

Oversight of previous work

The authors seem to have overlooked important recent literature on this topic including, for example, McNorton et al. (2022; doi:10.5194/acp-22-5961-2022) who used TROPOMI data to constrain methane emissions during COVID.

An egregious oversight that we have now addressed in the revised manuscript.

The authors don't seem to have reported uncertainties. It's clear what changes are actually substantial or within the noise. For example, the abstract lists changes of -3 Tg and -5 Tg as "substantial" in the abstract (Line 18). These don't seem particularly large. The text later claims that their work is within the uncertainty of another paper (Line 119), so it would be good to see uncertainties reported throughout.

Good point. We have now included the uncertainties for our regional changes in posterior emission estimates.

Error correlations

Where do the temporal and spatial prior error correlations come from? It seems that the authors use spatial correlation lengths of 300 km and 1 month. Are these important in the spatial patterns found here?

These are based on our previously published studies for which we show that using these correlation length scales are not important to the large-scale emission distribution we report in this manuscript.

Introduction

This reviewer is a bit confused by the list of citations in the intro. Specifically, Lines 34-35. The authors claim there is an intense debate on the role of fast growth in 2020 and 2021. They then claim that work has shown the importance of regional anomalies in the tropics. But many of these studies are from earlier than the time period being discussed.

The underlying reasons for these anomalous growth rates in 2020 and 2021 are currently subject to intense debate with some studies attributing most of the growth in 2020 to a reduction in the hydroxyl radical (OH) sink of methane due to global-scale reductions in nitrogen oxides due to pandemic-related industry shutdowns (Laughner et al. 2021). On the face of it, this appears to be a reasonable explanation, but recent studies have used satellite observations of atmospheric methane to reveal regional hotspots over the tropics that are responding to changes in climate and have global significance (Pandey et al. 2021; Lunt et al. 2019; 2021; Pandey et al. 2017; Feng et al. 2022; Palmer et al. 2021; Wilson et al. 2020).

The reviewer is correct. In the revised manuscript, we clarify the argument being outlined here. We focused on 2020 but we should have discussed the broader OH/emission argument, and the different data being used to make those points.

Correlative data

The authors show plots of the changes in correlative data, but don't show spatial correla- tions. In this reviewer's opinion, it would be helpful to show a map with the correlation between the emission anomalies and the correlative data. The manuscript currently requires the reader to make the connection themself.

We report correlations between different regions, as this reviewer has noted. We found that spatial distributions are more difficult to report based on two years of data, due to the coarse spatial resolution of our posterior emission estimates and due to regional time lags associated with rainfall and methane emissions. Consequently, we decided it would not be a useful addition to the manuscript. We explain in the revised manuscript why we do not include this figure.

Response to review comment 2

General:

Feng et al attempts to attribute the stunning increases in the methane growth rate from 2020-2021 using satellite observations of methane in conjunction with surface data. The subject is very timely and worth getting out into the community for further analysis and discussion. However, there are a number of issues that the authors need to address, which are incorporated into the annotated text of the paper. In particular, there are no uncertainty estimates of the fluxes, no real explanation of the Eastern African increase, and an unrealistic description of OH change. Spending more time on these elements will strengthen the paper and adding credibility to the results.

OH reduction is still challenging to quantify due to lack of (direct) observations. In the revised manuscript (Appendix D), we include additional inversion experiments based on different assumptions on 2020 OH reduction. In Appendix E, we estimate OH scaling factors (with the surface emissions of methane) from methane concentration observations. Our results all suggest that increased emissions in 2020 are primarily responsible for the large increase in the global atmospheric growth rate of methane.

Recent regional inversions based on both GOSAT and TROPOMI data also show large emission from Africa in 2020/2021 (not shown), and they are consistent with changes in hydrological data and land surface data. Further research is needed to understand the physical mechanisms responsible for the Eastern African emission changes, which is the outside the scope of this manuscript.

By how* much*?

By using a higher bias correction, we reduced the model overestimate compared to HIPPO aircraft data by about 5ppb in earlier years (2010-2011) prior to our experiment period.

For both CO2 and CH4, or just CH4?

It is just for CO_2 prior emission. For anthropogenic CH_4 emission, we have used EDGAR v4.32. We have restructured the paragraph so that the reader can easily find details about the CO2 and methane prior inventories.

Olsen and Randerson is a downscaling technique. What biospheric model did you use?

We used the results from the NASA CASA model (Randerson et al., 1996) which we now clarify.

A 5% reduction is not that conservative. The spatial pattern of OH reduction is quite important. Miyazaki et al, 2021 (Science Advances) shows that the global mean reduction in OH is about 4%. However, the reductions can be substantially higher, (>30%) locally. These will directly affect the methane inferences, especially over Europe and parts of Asia. Please review the paper and perform a sensitivity analysis closer to reported spatial changes in OH.

In our revised manuscript we performed two additional inversions with different (assumed) temporal and spatial patterns for the reduced OH, including

1) a higher (25%) OH reduction over China in March 2020.
2) global reduction pattern following observed ozone changes

We could expect, however, that the CO2 emission in 2020 and 2021 to be different than 2019. Not sure how this really addresses the OH issue. As noted next, the Miyazaki et al, 2021 provides a more observationally constrained pattern of OH.

Here we use CO_2 emission as proxy spatial pattern for the reduction of OH due to reduced human activity by COVID lockdown. In the revised manuscript we tested different assumptions for the OH reduction pattern and conducted a new inversion experiment by including OH scaling factors to the methane emission state vector to inferred from methane observations.

To echo our response to a similar comment from Reviewer 1, there are no effective constraints to provide accurate estimates for the spatial and temporal change in OH during the Covid lockdown.

Is this a correlation in flux or concentration space? If it is the former, where did 300km come from?

It is for surface emissions. We take this value from a previous study (Feng et al., 2022). It has no significant impact on the estimated global change of methane emissions between 2019 and 2020.

You should be able to calculate the simulated atmospheric CH4 growth rate for 2020 and 2021 from your inversion. You should add that to the table (1).

Good point. We estimate the global growth rate in 2019 is 6.7 ppb/yr, and 15.6 ppb/yr in 2020 from the simulation forced by posterior methane emissions inferred from GOSAT. There will be differences between the global growth rate inferred from GOSAT data and *in situ* data due to differences in geographical coverage.

The box-model approach carries its own assumptions, and can't be really used to validate the CH4 topdown estimates.

We use this model to show that based on a mass balance argument, satellite column XCH4 data show a larger net emission change between 2019 and 2020 compared to values inferred from the coarser in situ surface data.

Based upon NOx emission changes during 2020-2021, we would not expect CH4 lifetime to be fixed.

Please see our response to the previous question. Here we only the model to quantify the net emission change from mass balance. We agree that the methane lifetime may very well be different in 2020, and the implications of that our conclusions have been discussed in Appendix D and E.

There are no uncertainty estimates in this calculation. I can not assess whether these changes are tatistically significant or not. Uncertainty estimates need to be added to these flux estimates. These could be done through comparison with other inversions or independent observations.

We have added the uncertainties to regional fluxes.

It looks like the increases in SA in 2021 are an acceleration of the 2020 increases. What is going on there?

It is an interesting observation. Currently, we do not have sufficient independent data to explain this change.

How is anomalous defined here? The temperature variations need to be calculated relative to the long term temperature variability to assess whether they are not within climatology.

Those anomalies have been calculated based on the mean between 2010-2021 when GRACE data are available.

Please put plots of those regressions in the supplemental.

Good suggestion. We add the plot Figure A6 for correlation between monthly flux anomaly and GRACE LWE anomaly during 2018-2021.

Cooper didn't discuss NOx emissions, only concentrations. The impact on OH would need to be calculated separately and would be affected by other reactive species, e.g., ozone.

The reviewer is right. We have clarified this point and the impact on OH being more complicated.

It's hard to see the differences in a spatial plot. Please make a difference plot using the regions used in Fig. For 2020 and 2021 between the proxy retrieval and the simultaneous estimate.

We have added a new plot (Figure A7) in the revised manuscript, following this suggestion

The implication of East Africa as the single largest driver of the methane growth rate is puzzling. Yes, there is a correlation with water anomalies, but it's not the largest driver of methane in driver. A more plausible set of physical mechanisms needs to be proposed for why we could expect this region as a dominant driver.

This result is consistent with (and builds on) a number of preceding studies that have shown the relationship with rainfall, river flow down the Nile, and changes in the extent of the Sudd wetland. We have added Figure A6 that shows the strong relationship between liquid water equivalent anomalies inferred from GRACE and methane flux anomalies over the region. High resolution inversions enabled by TROPOMI have also highlighted that South Sudan is a globally significant source of atmospheric methane.

This is not clear to me at all that this is a valid assumption. The OH responses will depend on the regional NOx emission reductions and the timing of those reductions, in part because it will also affect ozone production. There are better sources for OH than this crude assumption.

We have now included an additional experiment that uses a different assumption to define the OH reduction pattern and have also included an OH inversion that infers scaling factors and methane emission estimates from atmospheric methane data. Our results all confirm the dominant role of increased emissions in explaining observed global growth of atmospheric methane.

References

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