

## Response to review comment 1

We thank the reviewer for their feedback. We respond below to each individual comment raised by this reviewer.

*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. At this time we do not have access to NOAA data for 2021.

We have also evaluated our posterior emissions using independent TCCON XCH<sub>4</sub> measurements for 2020 and the first half of 2021 (not shown). We find that the annual mean model minus TCCON XCH<sub>4</sub> values for 2020 are within 10 ppb for 15 (out of 18) sites with standard deviation between 5 and 15 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. We will accordingly update the text.

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 sufficient 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.*

An alternative way would indeed be to include emissions of methane and OH in the state vector but as the reviewer will be aware there are also gross assumptions associated with this approach and the resulting posterior results would not provide a "straightforward assessment" due to the limited information content of the satellite and *in situ* atmospheric methane data. Inversions that also include the estimation of OH, quantify OH values on global or hemispheric scales and therefore will be unable to identify OH hotspots. The ideal approach would be to include the methane emissions in a state vector as part of a full-chemistry data assimilation system that takes into account 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).

The approach 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 feel this is a transparent approach that is

easy to understand. We will include a discussion of this caveat in the revised manuscript. 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  $2^\circ \times 2.5^\circ$  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% 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 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. We will make this point clearer in the revised manuscript. 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 will add 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 NO<sub>x</sub> 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 will be 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). Since publishing his 2021 paper, Dr Miyazaki has re-run his calculations with coincident changes in hydrocarbons (private communication) and we are currently discussing how this information could potentially inform our results.

We did a number of simulation runs 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<sub>2</sub> emissions, resulting in a distribution similar to Miyazaki et al, 2021. An effective approach to identify regions affected by the shutdowns. 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. We will add a broader discussion of this kind of OH perturbation to the revised manuscript.

While we are happy to explore different OH perturbations, 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 will add 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 CO<sub>2</sub> concentrations. However, it seems like the authors use CO<sub>2</sub> simulations with monthly emissions through 2019. Therefore the CO<sub>2</sub> could lead to a bias in their methane concentrations during COVID due to the reduction in CO<sub>2</sub> emissions. This would be most pronounced in urban areas.*

As described in Parker et al., 2020, the GOSAT data use an ensemble of CO<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> values, an inversion is performed that directly uses the XCH<sub>4</sub>/XCO<sub>2</sub> ratio, without relying (explicitly or implicitly) on the model CO<sub>2</sub>. We will clarify this point in the revised manuscript.

*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 XCH<sub>4</sub> data (Line 90) but the main inversions also seem to use proxy GOSAT data.*

We use two approaches: 1) the proxy ratio (XCH<sub>4</sub>/XCO<sub>2</sub>) directly without need to assume a prior model for XCO<sub>2</sub> to multiply out to obtain XCH<sub>4</sub>, which is described in Feng et al, 2022; and 2) the proxy methane XCH<sub>4</sub>. We find both approaches lead to consistent results. We will amend the text to make this point clearer.

## *4 Minor Comments:*

### *4.1 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 will address in the revised manuscript. We will also provide a narrative about the comparison of results.

*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.

### *4.3 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.

#### 4.4 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 will broaden 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.

#### 4.5 Correlative data

*The authors show plots of the changes in correlative data, but don't show spatial correlations. 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 themselves.*

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 will explain in the revised manuscript why we do not include this figure.

## References

Feng, L., Palmer, P.I., Zhu, S. *et al.* Tropical methane emissions explain large fraction of recent changes in global atmospheric methane growth rate. *Nature Comm* **13**, 1378 (2022). <https://doi.org/10.1038/s41467-022-28989-z>

Lunt, M. F., Palmer, P. I., Feng, L., Taylor, C. M., Boesch, H., and Parker, R. J.: An increase in methane emissions from tropical Africa between 2010 and 2016 inferred from satellite data, *Atmos. Chem. Phys.*, 19, 14721–14740, <https://doi.org/10.5194/acp-19-14721-2019>, 2019.

Lunt, M. F., P. I. Palmer et al, Recent rain-fed pulses of methane from East Africa during 2018-2019 contributed atmospheric growth rates, 16(2), *Env. Res. Lett.*, <https://doi.org/10.1088/1748-9326/abd8fa>, 2021.

Sun, W., Zhu, L., De Smedt, I., Bai, B., Pu, D., Chen, Y., et al. (2021). Global significant changes in formaldehyde (HCHO) columns observed from space at the early stage of the COVID-19 pandemic. *Geophysical Research Letters*, 48, e2020GL091265. <https://doi.org/10.1029/2020GL091265>

Wilson, C., Chipperfield, M. P., Gloor, M., Parker, R. J., Boesch, H., McNorton, J., Gatti, L. V., Miller, J. B., Basso, L. S., and Monks, S. A.: Large and increasing methane emissions from eastern Amazonia derived from satellite data, 2010–2018, *Atmos. Chem. Phys.*, 21, 10643–10669, <https://doi.org/10.5194/acp-21-10643-2021>, 2021.