Reply to Review 1

We thank the reviewer for the constructive comments. Reviewer comments are provided below in black with our responses in blue.

General comments

The results in this version of the manuscript are better explained and should be published; however, there are many confusing word choices and comparison choices still present in the document. I therefore recommend that some of the more experienced authors in this manuscript carefully review this next version so that the next iteration is more likely to get through reviewer and editorial review.

In addition, as a reader, Im still struggling to restate the primary technical and scientific conclusions of the paper; I get that this is challenging as there are several (different types of) results described in this manuscript and summarized in the abstract…. Note that the short summary does a better job of summarizing results than the abstract so perhaps go over with your colleagues what you think the main results are and then come up with a succinct set of conclusions that can be stated in the abstract?

We have made some minor changes to the abstract for clarification and in response to other comments. After careful re-review we find that it is sufficiently clear and communicates our main findings well in its current form.

Specific comments

1a) Abstract, paper, and conclusion about estimating OH effect on methane emissions: I don't think you mean "cannot be simultaneously resolved" in the abstract but instead that you can estimate OH to XXX percent. For example, Im pretty sure you can falsify the hypothesis that the lifetime of CH_4 (via OH) is greater than 100 year (or alternatively less than 1) using TROPOMI methane. Instead the paper simply states that you cannot easily choose between the fixed versus variable OH results so you went with the fixed because it agrees better with aircraft… this is not a bad argument but is very different from saying that TROPOMI alone cannot resolve the effect of OH on methane. Changes to the language are therefore needed to reflect what you actually did versus what you think is happening (error bars on the OH estimate would help here if you can provide them!)

We have revised the wording on this point in the abstract as requested.

1b) Note that the methane life time (or alternatively OH abundance) is also estimated in the cited manuscripts but there is no comparison to the estimated uncertainties in these manuscripts. As you do not report an uncertainty for the effect of OH on methane perhaps add some additional relevant discussion comparing your results to these other papers.

Instead of listing individual uncertainty estimates for every cited OH paper in Section 3, we give the range in derived global-average OH concentrations across those studies. This provides another uncertainty metric and makes the point that our two candidate OH fields both fall within a viable range based on prior studies of the global OH budget. Adding individual uncertainty estimates for the cited papers would not alter that conclusion.

2a) Abstract: Need error bars on results, e.g. 571 +/- ???, 20 +/- ???

Added as requested.

2b) Abstract line 25: don't use the word "global upward" and "India / Southeast Asia" in the same sentence as this is confusing.

This sentence has been revised.

3) Abstract and general text. Now that I better understand the downscaling approach , I concur that it is novel and should be published, However, as discussed in Liu et al. 2021 and other TROPOMI papers, variations in albedo can easily alias into the (downscaled) emissions. Some discussion is needed ion how you determine whether the downscaled emissions represent a real source versus a radiative artifact. Also, you could emphasize that these systematic errors are somewhat mitigated with the downscaling approach by weighting both the prior and conserving total emissions at the 2x2.5 flux estimate resolution.

In our view this point is already addressed in the paper in the following sections:

"We use the SRON corrected retrieval described in Lorente et al. (2021), which is based on the S5P-RemoTeC full-physics algorithm with albedo correction and updated regularization scheme, spectroscopic information, and surface treatment. This updated algorithm mitigates the albedo bias that affected earlier versions (Qu et al., 2021). Relative to the albedo-corrected product, the prior TROPOMI version exhibits high biases over North Africa, the Middle East, and the western US, and low biases over Amazonia, the eastern US, central Africa, and eastern China (Lorente et al., 2021)."

"We omit high-latitude (>60°) observations and require quality filter QA > 0.5 (Sentinel-5 Precursor/TROPOMI Level 2 Product User Manual: Methane, 2022) to avoid errors associated with high solar or viewing zenith angles, low surface albedo, excessive aerosol loading, clouds, terrain roughness, and measurement noise (Lorente et al., 2021)."

4) Section 2.6 Line 250: I suggest that the first sentence in this section adds some specificity to the downscaling approach, e.g. "We use a combination of the TROPOMI column enhancements and a priori to partition fluxes derived at 2.5x2 (lon/lat) degrees to 0.1 x 0.1 degrees resolution."

This section has been revised accordingly.

"We present here a new method to spatially downscale the satellite-derived emissions from 2° × 2.5° to 0.1° × 0.1° for potential use in models. The downscaling, which combines information from the TROPOMI column enhancements, the prior emission estimates, and their uncertainties, is necessitated by the fact that the current GEOS-Chem adjoint model does not have global simulation capability at finer than 2° × 2.5° resolution."

5) Abstract and Line 570: There are a couple of places in the text where you talk about the emissions magnitude but then you reference a trend… this type of comparison is very confusing.

This comparison approach is even more confusing in line 570 where you are saying a trend cannot fully explain the TROPOMI emissions. Compare mean to mean, not trend to mean.

We thank the reviewer for pointing this out. In many cases our prior emissions are based by necessity on bottom-up inventories for previous years. A simple prior-to-posterior comparison therefore neglects any emission trend that is known or expected to have occurred between the time of the inventory and the time of our inversion. We therefore compare the derived emissions to the prior

value in the context of any trend that is estimated to have occurred in the interim. We have revised the text in multiple places for better clarity on this point.

6) Line 576 units?

Revised accordingly.

7) conclusion on wetlands.. Is the 149 prior estimate for wetlands from Ma et al.? If so cite reference and note that this is informed from GOSAT data

The 149 prior estimate is from Bloom et al. (2017) and Saunois et al. (2020). We have cited these references in section 2.3.

8) Im still a bit confused about one of Monsoon conclusions. The results imply that not including the monsoon results in an underestimate of the emissions. However, is this underestimate relative to a yearly mean (i.e. yearly mean based on monthly emissions is higher than yearly mean that is generated by estimating yearly emissions total such as in the Qu et al. and Worden et al. papers). Or does it mean that emissions are underestimated during peak rainfall?

The emissions are underestimated during peak rainfall, as follows:

"Approximately 80% of India's annual rain falls during the summer monsoon (IPCC, 2021), and across this Jul-Oct season we find that methane emissions from the India and Southeast Asia boxes in Figure 2b are underestimated by 37 (15–45)%."

Reply to Review 2

We thank the reviewer for the constructive comments. Reviewer comments are provided below in black with our responses in blue.

The authors have added more caveats about their results and clarified ambiguous descriptions of their method. I wish the authors had put more time into answering my concern with the downscaler and analytically propagated their assumptions to the final products (for example, what if X and Y prior emissions are used?). I still think the robustness of the downscaler wasn't proven to me.

As explained in our earlier reply and in the manuscript, we have in fact carried out a thorough evaluation of the downscaling method. This includes dedicated Observing System Simulation Experiments (OSSEs) performed both with and without transport error. Results demonstrate the robustness of the downscaling approach both in terms of the spatial fidelity of the solution and in terms of the derived flux magnitude. Specifically, the downscaled solution "yields a larger bias reduction (98% versus just 16%) and more accurate flux distribution (R = 0.70 versus 0.60) than the native fine-grid 4D-Var solution". Furthermore, "the downscaled OSSE solution reduces the prior bias by 17%–56% for sources exceeding 1000 kg CH_4/box/day (accounting for 99% of the domain-wide emissions) when not subject to transport error". The OSSE results also demonstrate that "in the presence of transport error, the downscaling method has limited success for the very largest sources (>2×10^5 kg/box/day), but nevertheless exhibits strong bias reduction (21%–50%) for sources between 1×10^3–2×10^5 kg/box/day (96% of domain-wide emissions)."

Some minor comments:

"Spatial emission biases" > what do you mean by that? Do you mean the prior knowledge used as pseudo-observations in the Bayesian framework is uncertain?

We have clarified this wording.

"Here, we instead combine multiple inversion frameworks" > multiple inversion frameworks are vague. You did not perform a multi-species/multi-sensor inversion. Please clarify.

We have revised this wording.

Also, we cannot validate emission with observations alone because other underlying issues in the model exist that are not fully constrained; the most direct way of validating an inversion is to compare the posterior flux to flux observations (i.e., eddy covariance matrices).

We agree that observed fluxes enable a direct comparison, but such observations are extremely sparse and subject to significant representation errors when comparing to a global model.

ATom employed the Airborne Tropospheric Hydrogen Oxides Sensor (ATHOS), with an estimated uncertainty of 0.018 ppt > 18 ppqv

We prefer to keep these units in their present form.

In the presence of transport error, the downscaling method has limited success for the very largest sources (>2×105 kg/box/day), but nevertheless exhibits strong bias reduction (21%– 50%) for sources between 1×103 –2×105 kg/box/day (96% of domain-wide emissions)." > awkward grammar.

Revised.

Please cite Zhang and Jacob et al., 2021 who found correlated information in the posterior covariance matrices of emissions and methane lifetime, suggesting that their segregation isn't possible by using CH4 observations alone.

We have added this citation.

A list of all relevant changes

Line 16-32: word change and clarification

Line 58: add citation

Line 109-110: clarification

Line 116-119: clarification

Line 220-223: clarification

Line 234: clarification

Line 252-254: clarification

Line 271-272: clarification

Line 309: word change

Line 312-313: word change

Line 431: revise typo

Line 435-436: word change

Line 440-442: clarification

Line 481: word change

Line 541-542: word change

Line 568-570: clarification

Line 575-577: clarification

Line 587: word change

Line 594: word change

Line 608-611: clarification

Line 633: word change

Line 638-639: word change