

We thank the South American researchers for their feedback on this manuscript. We addressed their specific comments in a separate reply, but we have also now added María Cazorla, Laura Dawidowski, and Sebastián Diez as co-authors. Their assistance in the revision process has helped strengthen the manuscript and add a much-needed local perspective, particularly with regard to our interpretation of inversion results.

We also thank the reviewers for their suggestions and comments on the manuscript. Please see below our responses to the comments. We have listed out the reviewer comments in black and the replies in blue.

### Reviewer #1

Hancock et al. present a comprehensive study that integrates satellite observations of CH<sub>4</sub> with an atmospheric transport model and prior emissions estimates to derive an optimal set of methane emissions for South American countries. This analysis further disaggregates the optimized emissions by sector and country, enhancing the atmospheric constraint on South American methane emissions. The topic is significant, and well-aligned with the scope of ACP. I am not an expert on inverse modeling but rather on in-situ measurements so I can't comment deeply on the mathematical aspects of the inversion method and learnt a lot. Below, I outline a few questions and suggestions.

**Line 93:** what is the retrieval success rate specifically for South America?

The retrieval success rate for South America is 28.4%. We now note this in the text (lines 99-100).

**Regarding Fig. 1:** How would the seasonal variability of methane concentration affect these emission estimates? A spatial plot of the total number of samplings in each season during 2021 might be useful to include in the supplement. Is it possible to extend this inverse modeling setup to estimate total methane emissions on a monthly scale?

Thank you for this suggestion. Since we have higher observation density in the dry season (June-September), our emissions estimates are more heavily based on observations during this period. We have now included a figure in the supplement showing seasonal TROPOMI and GOSAT observation density (Figure S1). We also show in Figure S1 that the mean bias between GEOS-Chem and TROPOMI+GOSAT XCH<sub>4</sub> is lower in the posterior than in the prior in all seasons.

It is possible to use a similar inverse modeling setup to estimate methane emissions on a monthly scale. Varon et al. (2023) demonstrated this over the Permian Basin with weekly monitoring of emissions using TROPOMI. While looking at seasonal variability over South America would be very interesting, it would require redoing all of the  $0.25^\circ \times 0.3125^\circ$  perturbation simulations, representing over 100 thousand hours of computation time. We hope this can be a focus of future work.

**Line 115:** "There are few observations over the mountainous Andes, affecting much of Chile and Peru, so the inversion for those countries relies significantly on glint observations offshore and on observations of transported methane." How does this affect the uncertainties in estimating emissions for this area?

The smaller number of observations over these countries is reflected in their lower averaging kernel sensitivities shown in Table 2. We now emphasize this in the text (lines 420-421).

How does the inverse model handle temporal variability in emissions? While the model optimizes the overall magnitude and spatial patterns in emissions, does it also optimize seasonal or year-to-year variability? In other words, does the model assume that the temporal distribution of emissions is known or fixed according to the prior temporal distribution?

We optimize annual emissions such that the inversion assumes the temporal distribution of emissions follows that of the prior. The primary driver of seasonality in emissions is wetlands, and we test two different wetland prior inventories to characterize the uncertainty that comes from this, but we do not optimize the seasonality of emissions.

**Line 206:** "We use 600 Gaussian functions as state vector elements to balance aggregation and smoothing errors." While a reference is provided, a brief explanation of why 600 Gaussian functions are used would be helpful.

Thank you. We have added this to the manuscript (lines 223-225).

**Regarding Fig. 3:** The ratios of the posterior/prior emissions in Fig. 3c show values close to zero or over 2 in many areas (e.g., Bolivia and Argentina). Does this imply that the inversion zeroed or doubled emissions? If so, are the resulting emissions reasonable?

Thank you for this question. Yes, the ratios indicate what the prior emissions in a particular grid cell should be multiplied by to obtain the posterior emissions, so a ratio of 2 would mean the emissions are doubled and a ratio of 0.5 would mean the emissions are halved. We consider the resulting emissions to be reasonable, especially because the very large and very small ratios are generally limited to areas with low prior emissions and thus the magnitude of change in emissions is small.

Given the goal of the paper is to "evaluate the national inventories submitted to the United Nations Framework Convention on Climate Change (UNFCCC) under the Paris Agreement and to identify opportunities to improve countries' bottom-up reporting methods," including model-data comparisons against independent CH<sub>4</sub> observations is crucial for evaluating the inverse model. While there is a comparison with aircraft measurements in the Bolivian Amazon region, this is not enough. A supplement showing the bias of methane concentration in the prior and posterior run relative to in-situ observations in South America would be very informative.

Thank you for this suggestion. We have now included a comparison with the Amazon Tall Tower Observatory (ATTO) in Brazil (Figure 5).

**Minor Point: Line 98:** TCCON is first mentioned here. It should be referred to as the "Total Carbon Column Observing Network (TCCON)." Additionally, providing a brief explanation of TCCON would help readers unfamiliar with this field understand its purpose.

Thank you for this suggestion. We have added this to the manuscript (lines 104-105).

## Reviewer #2

This paper presents a comprehensive analysis of methane emissions across South America. By employing high-resolution satellite data from TROPOMI and GOSAT, the authors present a comprehensive and spatially detailed estimation of methane sources, mainly anthropogenic sources, which constitutes a significant contribution to the comprehension of regional methane budgets. However, there are a few aspects of the paper that could be addressed to yield a more robust and comprehensive analysis.

The integration of data from two complementary satellite instruments helps to improve estimates through the use of inverse modelling. Nevertheless, a validation with independent CH<sub>4</sub> observations available in South America would be beneficial for this study. For instance, the authors could undertake a comparison of the posterior mole fraction with data obtained from the ATTO tower in Brazil and vertical profiles in the Amazon region.

Thank you for the suggestion. We have now included a comparison with the ATTO tower in Brazil (Figure 5).

With regard to the regional budget, it would be beneficial to include a comparison with other previously published top-down estimates of total methane emissions for each country, such as those included in the Global Methane Budget.

The authors could additionally provide insight into the implications of their findings for policymaking strategies. This would be beneficial, as they evaluated national anthropogenic emissions inventories reported by individual countries to the UNFCCC. In particular, the authors could elucidate how these data could help local governments to mitigate methane emissions.

Thank you for this suggestion. We have included a regional comparison to estimates from the Global Methane Budget (Saunois et al. (2024)) and a global inversion by Worden et al. (2022) in the supplement (Figure S2).

We have changed the language of the paper to clarify that the intention of this work is not to evaluate countries' UNFCCC reports, but rather to begin to explore possible causes of the mismatch between top-down methane emissions estimates and bottom-up inventories that have been identified in global inversions (Worden et al., 2022). This work is only a starting point for addressing South American methane with satellite observations and our hope is that it can be followed up by country-specific studies working with local governments and scientists that will provide more policy-relevant results.

Specific comments are provided below.

Line 20: The term "correcting" may suggest that top-down estimates are inherently more accurate than bottom-up estimates, whereas both approaches are subject to their own sets of uncertainties. To prevent any potential misinterpretations, it would be helpful to use a term such as "adjusting" or "reconciling" when discussing the comparison or combination of these estimates. It is also important to discuss the limitations of both methods, top-down and bottom-up estimates, in the paper.

Thank you for this suggestion. It is common in the top-down literature to refer to 'correction' and correct (verb) as the change from the bottom-up prior, but this does not mean that the top-down results are correct. We have changed the language and added additional discussion of the uncertainty of top-down methods throughout the text per your suggestions.

Lines 118-119: state that satellite observations are distributed throughout the year, but are most dense during the southern hemisphere dry season (June-September). How the lower density of observations during the wet season in comparison with the dry season could affect the posterior estimates. This is particularly relevant given that this region has extensive wetland areas, where the highest emissions are expected during the wet season.

Thank you for this comment. Since we have higher observation density in the dry season (June-September), our emissions estimates are more heavily based on observations during this period. We have now included a figure in the supplement showing seasonal TROPOMI and GOSAT observation density (Figure S1). We also show in Figure S1 that the mean bias between GEOS-Chem and TROPOMI+GOSAT XCH<sub>4</sub> is lower in the posterior than in the prior in all seasons.

Figure 1 illustrates the annual mean 2021 dry-column methane mixing ratios (XCH<sub>4</sub>) after subtraction of background, clearly demonstrating the absence of TROPOMI data in the Amazon region, which is compensated by GOSAT observations. However, an examination of the plot of the number of observations shows that there are fewer GOSAT observations during the months of April to June in comparison with other months. Please describe the extent of data coverage for South America during this period, with particular attention to the Amazon region. It would be beneficial to have a map as supplementary material that includes both TROPOMI and GOSAT dry-column methane mixing ratios (XCH<sub>4</sub>) for the initial period of the year, particularly April to June.

Thank you. As you suggest, we have now included a figure in the supplement showing monthly TROPOMI and GOSAT observation density (Figure S1).

Lines 196-200: What is the lifetime of methane considering all the sinks, including oxidation by hydroxyl (OH) radicals and tropospheric chlorine (Cl), oxidation in the stratosphere, and uptake by soils?

Thank you for this question. The lifetime is  $9.1 \pm 0.9$  years (Szopa et al., 2021), which we state on line 45 of the manuscript. We have also added this on line 227 for clarity.

Line 328: "Most of that increase is from anthropogenic emissions". Does this imply that the prior estimated wetland emissions for the South American region are consistent with the atmospheric

measurements? Alternatively, could the posterior wetland fluxes be more dependent on the prior estimates due to the limited observations in the Amazon region (which has larger methane emissions, as illustrated in Figure 2), as reflected in the low averaging kernel sensitivities? It would be beneficial to conduct a comparison with independent atmospheric observations to evaluate the posterior estimates.

That sentence does not imply that the prior wetland emissions are consistent with the satellite observations, but rather that the upward adjustment to wetland emissions is smaller than the total upward adjustment to anthropogenic emissions. This is a great point that this could be dependent on the lower averaging kernel sensitivities. In section 3.2, we describe the adjustments to wetland emissions in greater detail. Despite limited observations over the Amazon, we obtain an averaging kernel sensitivity of 0.74 over the region, indicating that our estimate is more informed by the observations than by the prior estimate.