

## Referee #1

## We thank the Dr Boone for insightful comments. Our replies are in the red coloured text.

The authors have answered all my comments to the previous version of the paper in a sufficient way. I only have a few technical comments to the revised version of the paper.

1. There are still several sentences (esp. in the newly added text) where e.g. 'the' is missing. I assume this will be handled during language editing.

## Yes. We have tried to edit the text further but ESSD does also have dedicated team for style/grammar editing.

2. l. 51: 'Note that SPARC data uses earlier (v3.6) version of the ACE-FTS data' → 'Note that SPARC data use an earlier version (v3.6) of the ACE-FTS data.'

## Done

3. l. 52: 'data contains' → 'data contain'

## Done

4. l. 215: 'We have added discussion in a revised manuscript' This sentence should be deleted, or do you refer to another manuscript?

## Done, we have removed the sentence.

5. l 316/317: 'Though exact causes of unusually low CH<sub>4</sub> values in S-MIPAS-CH<sub>4</sub> and S-ACE-CH<sub>4</sub> data files are unclear but might be associated with the downwards transport of CH<sub>4</sub> poor air...' Unclear sentence, probably 'Though' should be deleted.

## Done, revised as "A possible explanation might be the absence of negative data points seen in ACE data (due to enhanced winter-time downward transport of CH<sub>4</sub>-poor air) which are excluded in the XGBoost training step."

## Referee #2: (Chris Boone)

The updated version of the manuscript shows improvement. I have no major concerns remaining, just some minor comments.

## We thank the Dr Boone for insightful comments. Our replies are in the red coloured text.

>Line 245: Additionally, the onion peeling algorithm used for solar occultation measurements assumes observations at different tangent height are independent, hence retrieved profiles show larger fluctuations.

ACE-FTS analysis does not use onion peeling. The observational results will have 'random errors' contributing to the variability regardless of the analysis approach employed, for example from the impact of measurement noise in the analysis. The model data has no such contribution to the variability.

## Modified to say "some occultation instruments such as SAGE/ HALOE"

> Line 289: As the ACE-FTS retrieval algorithm uses multiple micro-windows, a seasonal variation in vertical structure of the atmosphere alters spacing between tangent heights.

The spacing between tangent heights has no connection to the micro-windows. Spacing between tangent heights for ACE-FTS measurements exhibits a pseudo-seasonal variation as the occultation measurement geometry changes systematically over the course of a year.

## Thank you, we have removed the discussion about the micro-windows in the retrieval. Revised as "As the forward model used in the ACE-FTS retrieval algorithm needs spectra on fixed height levels, a seasonal variation in vertical structure of the atmosphere alters the spacing between tangent heights."

>Line 166: We are aware that some tracers are correlated as all the variables are from a TOMCAT simulation (or forcing meteorology) ...

Note that all the tracers used are also measured by ACE. It would be possible to separately evaluate each tracer, but that is presumably beyond the scope of this paper.

## Yes, we agree that it is beyond the scope for this paper.

## Typos:

>Line 51: Note that SPARC data uses earlier (v3.6) version of the ACE-FTS data  
...uses an earlier...

## Done

>Line 210: The time term (date) term...

'Term' is used twice

## Done

>Line 258: Figures s5 and S6

One lower case s (s5) and one upper case (S6)

## Done

>Line 284: a seasonal minima occurs  
“minima” is plural.

## Done

>Line 361: On the other hand, differences at 35 km in Figure 8 seem to be dominated by the QBO-induced meridional circulation patterns Baldwin et al. (2001), that are underestimated in TOMCAT. This sentence is not grammatically correct (perhaps ...reported by Baldwin et al. or ...detailed by Baldwin et al.).

## Modified as (e.g., Baldwin et al., 2001).

### Referee #3

On careful reading of the Conclusions, I find some confusion that may be just me or may be fixable with language:

## We thank the referee for his/her insightful comments and our replies are in the red coloured text.

L388: 'constraint' is odd, the stratospheric profile are a key component of the X-CH<sub>4</sub> that must be subtracted. I do not think of them as 'constraining'

## We think it is a constraint for the retrieval algorithm. We have modified the sentence to be "Also, accurate stratospheric CH<sub>4</sub> profiles are a valuable constraint for the retrieval of tropospheric methane using satellite instruments."

L390: likewise 'dynamical scheme' is strange. Numerics is also relevant. Why not just "dependent on the chemistry and transport processes in a particular model"

## Thank you for a useful suggestion. Revised the sentence as "Furthermore, although chemical models are able to simulate long-term profile data sets of these species, they are highly dependent on the representation of individual chemical and dynamical processes in a particular model."

L398: "vary under". Do you mean they have different values in different location-times because of the regimes? or that they have different variability?

## Sorry for the confusion. We have revised and divided in to two sentences as "Because atmospheric concentrations of CH<sub>4</sub> and N<sub>2</sub>O vary due to distinct dynamical and chemical processes in different regions, our approach involves dividing global measurements into five latitude-based categories. These categories include two for polar regions, two for mid-latitudes, and one for the tropics. We then proceed to derive regression parameters for each 1 km vertical grid spanning from 15 to 60 km within each of these latitude bins."

L408: "the drift in Aura-MLS" This does not make it clear that the 'drift' is a bias or error, can you make it clear that it is an error?

## Added a reference and reworded as "confirms the abnormal drift in Aura-MLS v4.2 N<sub>2</sub>O data (as used in SPARC data set) especially at lower latitudes and altitudes (e.g., Livesey et al., 2021).

L410: "near negligible trends ..." The MLS N<sub>2</sub>O data show a clear positive trend in the upper strat that is greater than the surface trend, that is why the lifetime is dropping. What is meant here?

## We agree that MLS data shows positive trends in the upper stratosphere (Prather et al., 2023) but Minganti et al. (2022) show almost negligible trends in ACE-FTS data. We revised the sentences as:

"A casual inspection of TCOM-CH<sub>4</sub> and TCOM-N<sub>2</sub>O plots also suggests that despite increasing surface values there are near-negligible long-term trends in the upper stratosphere/lower mesosphere which is consistent with Minganti et al. (2022). On the other hand, Prather et al. (2022) analysed MLS V5 N<sub>2</sub>O data to showed positive trends (up to 15%) in the tropical upper stratospheric N<sub>2</sub>O, though they do not find NO production rising at the similar rates. A possible explanation would be variations in stratospheric/mesospheric loss processes, probably determined by changes in the stratospheric

circulation, is reducing the lifetime of these GHGs. We aim to analyse these discrepancies in a future study.”