

Review of “Using machine-learning to construct TOMCAT model and occultation measurement-based stratospheric methane (TCOM-CH₄) and nitrous oxide (TCOM-N₂O) profile data sets” by Sandip Dhomse and Martyn Chipperfield.

Overall, this is a well written and well-considered construction of long-term data sets for two important atmospheric molecules. I just have a few concerns that should be relatively easily addressed.

We would like to thank Dr Boone (Reviewer #2) for his insightful comments on our manuscript. His feedback has been invaluable in helping us to improve the clarity, accuracy, and overall quality of the paper. Our replies are in blue italic text.

> Line 225: “Additionally, the onion peeling algorithm used for solar occultation measurements assumes observations at different tangent height are independent, hence retrieved profiles show larger fluctuations.”

HALOE used onion peeling in its retrieval, but ACE-FTS does not. That is not the reason the variability is so high here. In this latitude region, you will see effects from atmospheric descent (the entire profile sinks to lower altitude in the stratosphere) inside the polar vortex during the winter. This would account for the pronounced ‘bulge’ in the variability around 20-25 km relative to the “trop” set, for example, which is the only region here that does not include a contribution from atmospheric descent. Additionally, in the lower stratosphere, you will see variability in CH₄ from H₂O-related chemistry, but most of the large variability seen in the data around 25-30 km presumably results from the inclusion of profiles experiencing different degrees of atmospheric descent inside the polar vortex over the course of the winter. It is a real, physical variability, not a retrieval artefact.

Yes, we agree. We have removed the sentence about the retrieval errors and averaging kernels and focussed more on wintertime downward descent leading to seasonal changes in tangent height might be causing dynamically forced strong annual cycle. We also highlight differences between two retrieval algorithms.

> Line 231: “...somewhat larger differences for 2019-2021 time period is that there has been rapid increase in atmospheric CH₄ over the last few years (e.g. Nisbet et al., 2019).”

Note that all the evaluation period (2019-2021) CH₄ comparisons exhibit a bump around 35 km, where ACE-FTS results are slightly higher than the TCOM-CH₄ results. This appears to coincide with the observation of larger trends at higher altitudes in ACE-FTS results that seem to result from a less efficient conversion of CH₄ to H₂O in the middle stratosphere in recent years (which leads to higher levels of CH₄ in later years):

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Thank you very much for pointing this out. However, we don't believe that this conclusion from Fernando et al. could be linked to the biases seen at 35 km. We think TCOM biases are likely due to large dynamical variability, use of the measurements containing only positive values and limited spatial coverage in the tropics. Figure S2 clearly shows that R² values for TCOM-CH₄ in the tropics are almost constant (close to 0.5) from 25 to 50 km.

>Line 242: “...in percentage terms biases can reach up to 100% near 40 km as changes in the small values can translate into much larger changes in relative differences.”

In Figure 3, the shape of the percentage change looks quite similar to the HNO₃ contribution to the N₂O correction that is shown in Figure 1. The resemblance in shape looks even more pronounced for the SHmid case (Figures S3 and S9 in the supplementary file). To me, that

looks somewhat suspicious. Are you certain HNO₃ is functioning as intended in the analysis? It looks like it is introducing a large percentage difference for N₂O in the 2019-2021 evaluation period.

We have double-checked our code once again and the HNO₃ proxy is correctly incorporated. The HNO₃ importance being larger than other proxies most probably indicates that NO_y (HNO₃ being major contributor) species partitioning might be biased in TOMCAT.

> Similar to CH₄, a seasonal minimum occurs just after the break-up of Antarctic polar vortex (October), transporting N₂O-depleted air to lower altitudes. The seasonal minimum is not a consequence of the break-up of the polar vortex, it is the result of atmospheric descent within the polar vortex before it breaks up.

We agree; the paragraphs have been revised in the manuscript. However, the final vortex breakup only happens when there is major dynamical wave driving that suddenly strengthens stratospheric circulation. At the same time, this enhanced activity increases horizontal mixing as well (isentropic transport). So, major changes in the lower stratospheric CH₄ happen only after polar vortex breakup. In contrast descending air masses inside the polar vortex, affect only small part of the stratosphere.

>Line 262: "As the ACE-FTS retrieval algorithm uses multiple micro-windows, there may be a seasonal shift in averaging kernels causing fluctuations in the retrieved profiles."
ACE-FTS retrievals do not use averaging kernels. There is a seasonal variation in the spacing between tangent heights, and VMR profile variability could increase when tangent heights get very close together. When you get VMR values close to zero, it is normal to get negative values for an individual occultation. Note that excluding negative values and keeping only positive ones will actually introduce an artificial positive bias into averaged results.

We apologize for any confusion caused by our previous statement. We will add a sentence explaining the issue, as well as the possibility of positive biases in TCOM data because we only use positive values. We also plan to use both positive and negative values from ACE data during the construction of a new version of TCOM data.

>Line 288: "The exact causes of unusually low CH₄ values in S-MIPAS-CH₄ and S-ACE-CH₄ data files are unclear."
This is presumably another instance of atmospheric descent, with the descent signature in the data extending lower in altitude than in the model.

We will reiterate the role of the descending air masses once again.

> Line 314: "...the latitude slice indicates significant variations between two".
...between the two.

Done