Thank you very much for helping us to increase the quality of this article. Here below are the answers to your remarks.

A few specific comments:

P2,L23: "thermodynamic structure and atmospheric composition" **Done**

P6L5-7: "DFB" = distributed feedback; "ICL" = interband cascade laser Done

P6,L13: "Judson" is "Teledyne Judson Technologies"; PTU should be explained **Done**

P6,L14: "PTU" = pressure, temperature and humidity **Done**

P6,L23: "sensitivity" represents a signal/analyte quantity--do you mean "detection limit" (over what time?)? Table 1 shows values for both CH4 and CO2 in 2015 while P5, L21-22 states that in 2015 AMULSE was a single gas CH4 methane instrument and the dual-gas version was introduced in 2016. Table captions go at the top of the table, figure captions below.

I mean sensitivity. Indeed, the dual-gas version was introduced in 2016. Before that we had two instruments, a CO2 single gas version and a CH4 single gas version. I added an explanation (with two single-gas versions CO2 and CH4) in the caption and put it at the top.

Figure 1: comment in text on CO2 drift from early to late part of time series. Average of the difference does not capture--average absolute difference would be a better metric in this case. Stating the std dev as you have is valuable. Greater color difference between traces would be helpful.

This is the absolute difference I use, I'll notify it in the text and I changed the color of the curves (changed in main text).

P11,L7: "CO2 near the surface", "at night"; one reviewer suggested a reference for this. The artifact in H2O due to outgassing on ascent is mentioned, which explains the non-physical values shown in Fig. 5c, but why aren't descent data shown--the text mentions valid data then. A temperature profile plot would be a useful addition to Fig 5 and help with identification of the tropopause.

I added a publication for the day-night cycle of atmospheric CO2 concentration (due to photosynthesis) \citep{Schmidt2014} add in bibtex. I also added the temperature to better identify the tropopause level. I also changed the description of the figure.

P15,L9: interpolate or average? Doesn't the in situ data have higher vertical resolution so that you would need to average the data over the depth of a model layer? Fig 7--would probably be better to put the reference trace on top, so that it is clearer that the profile in the shaded (non-measured) region is not the from the sonde.

That is indeed what was done in this case. The in-situ profiles were interpolated to the fixed pressure levels by averaging the layers centred on these levels.

Modification in the text:

«... ,we have interpolated the in-situ profiles on the 101 fixed pressure levels by making layer averages centred on these levels. \gg

This is an interesting idea for Figure 7. It has been modified accordingly.

P16,L11: "red"? Red might actually be better

Also good idea, Figure 8 has been modified.

P16,L13: "This" here is confusing since it naturally refers to the "good agreement" when you are addressing the observed differences in the window regions. "The difference seen between the simulated IASI and CrIS window channels is due to the temporal and spatial differences between the selected pixels."

Corrected in the document with this suggestion.

« The difference seen between the simulated IASI and CrIS window channels is due to the temporal and spatial differences between the selected pixels, which implies a different skin temperature used in RTTOV »

P17,L7+: as a reviewer asked/suggested, explain 'EXP'. You did so in your response, but not in the manuscript.

Added in the paper :

« Figure 5 shows the difference between simulated brightness temperature (BT^S) with O_3 in purple (resp. CO_2 in red or CH_4 in orange) *in situ* measurement profiles (X^M) and simulated brightness temperature with O_3 (resp. CO_2 or CH_4) reference profiles (X^R) for *in situ* sounding from 2018-04-17 at 10 UTC for all channels contained in the IASI (a) and CrIS (b) spectrum, where EXP means experiment (for example, EXPO3 means experiment for ozone case, etc.), such as: »

P18: adjust colors in Fig. 9 to be more distinct. Clearer structure in the figure caption. Why is SONDE capitalized?

Figure 9 has been reconstructed. SONDE is in capital to mark the comparison to AMULSE

P18,L8: What "specific case"? "For the case of 2018-04-07, it can be seen in Fig 9 that..."

Changed in the text:

« For the case of 2018-04-07, it can be seen in Figure 9 that our static reference profiles of carbon dioxide and methane lead to realistic simulations, as for *in situ* profiles »

P20,L24: reference for the higher difficulty in CH4 simulation in CTMs (just source term or chemistry as well?) and the major scientific questions regarding CH4 trends.

Specific references to these issues have been added in this paper.

Archibald et al., 2020 « Description and evaluation of the UKCA stratosphere--troposphere chemistry scheme (StratTrop vn 1.0) implemented in UKESM1 »

Houweling et al., 2017 « Global inverse modeling of CH4 sources and sinks: an overview of methods »

Naik et al., 2013 « Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) »

Non-public comments to the Author:

There are some usage and construction issues that make the manuscript a little difficult to parse and understand in places. I believe that much of this will be addressed during the copyediting state, and I encourage you to work with the Copernicus copyediting professionals at that time. One high profile example is the usage "its interest in" in the title and elsewhere, which implies that AMULSE is interested in atmospheric applications. A better construction might be more simply "and application for satellite retrieval validation".

I agree, I changed the title.