Response to the reviewers on the manuscript "Methane retrieved from TROPOMI: improvement of the data product and validation of the first two years of measurements" by Alba Lorente et al.

The authors would like to thank the reviewers for their thoughtful and helpful comments and suggestions. Below are the comments by the reviewers in blue and replies in black. Any modification made to the text has been underlined. The line and page numbers correspond to the version of the manuscript available for online discussion.

Reviewer 1

Comment C 1.1 — Page 1, line 2: I recommend to add "and sampling" after "spatial resolution" as TROPOMI has a similar spatial resolution as GOSAT but much denser spatial sampling.

Reply: Added. We have also been more specific on the sampling technique of GOSAT on page 6, line 4.

Comment C 1.2 — Page 1, line 5: "The updated TROPOMI CH4 product...": If possible, please add version number. Does this product exist, i.e., is it available for interested users? If not, then please write "The updated TROPOMI CH4 retrieval algorithm...".

Reply: It is an existing product and it is publicly available through the ftp specified in the "Data availability" section. However, as we do not want to confuse the reader with version numbers and detailed specifications about the product in the abstract, we have modified the sentence to "The updated retrieval algorithm..." as the features that follow in that sentence refer to the algorithm itself.

Comment C 1.3 — Page 2, line 24: Barre et al., 2020: Missing in section "References". Please add. Please add that there is (at least) one other product as described in Schneising et al., 2019, and Schneising et al., 2020. These publications need to be cited (see References below) and the results shown in Schneising et al., 2020, need to be mentioned, especially those related to the Permian basin (see line 22).

Reply: We have added Barre et al. (2020) to the reference list; this was forgotten because when preparing this manuscript it was still under discussion in ACPD.

We agree that the WFM-DOAS TROPOMI product (Schneising et al., 2019) should be mentioned. We have mentioned it in Section 2.1 (TROPOMI CH_4 retrieval algorithm), page 4, line 27. We think this location fits better as it is here where the retrieval algorithm is presented. The added

text is: "Another scientific retrieval algorithm using the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) method to retrieve CO and CH_4 from TROPOMI was presented by Schneising et al. (2019). Comparison of both retrieval approaches is foreseen as part of ongoing verification activities."

We have added Schneising et al. (2020) when we refer in the text to the studies of the Permian basin. We do not go into the details of neither Schneising et al. (2020) nor Zhang et al. (2020) as the aim of this paragraph is to highlight some of the studies that have successfully used TROPOMI XCH_4 data to derive emissions.

Comment C 1.4 — Page 4, line 15, and Eq. (4): Instrument noise is not the only contributor to "XCH4 random errors", i.e., precision, as also other instrumental (e.g., inhomogeneous scene illumination) and retrieval errors (e.g., unconsidered variability of albedo and aerosols) may contribute. I suggest to add this limitation or, alternative, state that Eq. (4) is the definition of precision as used for this manuscript.

We acknowledge that there are other contributions to the random error besides the measurement noise. So it is true that Eq. 4 is the definition of the precision given in the product and so as used for this manuscript. As suggested by the reviewer, we explicitly mention this. "The precision σ_{XCH_4} available in the data product is defined as the standard deviation of the retrieval noise".

Comment C 1.5 — Page 4, line 21: "In cases when VIIRS data is not available, we use a back-up...": Does this happen? If yes, I would expect that this results in inconsistencies. Please add more information.

Reply: Data from VIIRS is hardly ever not available, so this does not happen very often. VIIRS data used in the TROPOMI XCH_4 retrieval is processed operationally by the S5P-NPP cloud processor. If due to any circumstance the processing of the VIIRS data fails or it is delayed, we use this filtering as a back up option. The XCH_4 data is flagged accordingly (qa value downgraded to 0.4) to avoid any possible inconsistencies as mentioned by the reviewer. From all the orbits processed operationally since the beginning of the mission, for less than 1% the processing of VIIRS data was not nominal in the CH_4 retrieval.

We added the following to clarify this point: "In less than 1% of the cases when VIIRS data is not available, we use a back-up filter based on a non-scattering CH_4 retrieval from the weak and strong absorption bands (Hu et al., 2016). These cases are flagged accordingly by the quality value indicator."

Comment C 1.6 — Page 4, line 27 following: "This updated retrieval algorithm is referred to as the beta version of the TROPOMI XCH4 data product." Sentence not OK. An algorithm is not a data product.

Reply: We agree with the reviewer about the misleading terminology used here. We have modified the text as follows in page 4, line 27: "The TROPOMI XCH_4 scientific data product from SRON retrieved with the updated algorithm serves as a beta version of the operational processing."

Following this comment, we have further clarified at the beginning of Sect. 3 (page 6, line 20), removing the reference to version 1.3.0 that will eventually correspond to the future operational update but this is not certain as of now: "The TROPOMI XCH_4 scientific data product from SRON retrieved with the updated algorithm will be suggested for use in the operational processing in the next processor update.."

Comment C 1.7 — Below Tab. 1: "*For the Lauder station the ll instrument was replaced on October 2018 to ll.". ll replaced by ll?

Reply: We thank the referee for spotting the typo. The instrument "ll" (Sherlock et al., 2017) was replaced by "lr" (Pollard et al., 2019). We have corrected this.

Comment C 1.8 — Page 5, line 9: If the TROPOMI data are averaged daily then I assume that the TROPOMI XCH_4 averaging kernels have not been considered for the validation. Please add more info on this aspect.

Reply: The total column averaging kernel can only be used when CH_4 profile measurements with a high vertical resolution would be available for validation. However, the measurements from the TCCON network only provide total column integrated measurements which hampers the application of the averaging kernels.

Comment C 1.9 — Page 6, line 17: "both retrievals performed similarly": With respect to what? Likely not w.r.t. yield as number of data points in proxy product is much higher. Please refine the statement.

Reply: We agree with the reviewer that we should be more specific in this statement. We modify the text for that purpose: "...both retrievals performed similarly with respect to bias variability and precision when validating the retrieved XCH_4 with ground-based TCCON measurements. This study also concluded that both methods can retrieve XCH_4 in aerosol loaded scenes with retrieval errors of less than 1%."

Comment C 1.10 — Page 7, line 4: "and that retrieved aerosol parameters have realistic distributions". This is a strong (but unproven) statement. It needs to be shown in this paper that this is true.

Reply: We agree with the reviewer that this statement needs clarification. First, to avoid misinterpretation of the output of the retrieval to which we refer as "retrieved aerosols parameters",

we change the reference to them in the manuscript to "scattering parameters" instead of "aerosol parameters", and add the prefix *effective* ("effective aerosol distribution height", "effective size parameter" and "effective aerosol column"). With effective we want to highlight that these retrieved parameters are auxiliary parameters that characterize the scattering properties of the atmosphere in the radiative transfer model in the retrieval for which the target is XCH_4. The aerosol parameters are only effective ones but follow a distribution that we would expect, and that is what we meant by realistic distributions. We have modified the sentence in page 7, line 4:"[...] retrieved scattering parameters follow a distribution that we would expect".

Comment C 1.11 — Page 7, line 12: "19.7 ppb to 24.5 ppb": What does this mean? Is it a min to max range?

Reply: It refers to the reduction on the standard deviation of the differences mentioned at the beginning of the sentence. We add 'from', and correct the order because the reduction is from 24.5 to 19.7 ppb. Furthermore, there was a typo and 24.5 ppb is 21.5 ppb, which matches the 9% reduction specified in that same sentence.

Comment C 1.12 — Page 8, 6-7: "we have decided to use the SEOM-IAS spectroscopy database." I am not convinced. Was this a "political" decision? I conclude from Tab. 2 that HITRAN 2008 (used so far) is better. Is a slightly better fit quality (which can have many reasons in addition to spectroscopy) really a good argument if bias and scatter are getting larger?

Reply: We acknowledge that the text can be somewhat misleading. The "slightly" better fit quality refers to the results when looking only to retrievals around the TCCON stations. On a global scale (page 7, line 30) "we see that both the RMS and χ^2 improve significantly when using the SEOM-IAS database, with HITRAN 2008 giving the worst fitting results". The prove of this statement is not visually shown in the manuscript, but we have added the following to the text as suggested by Referee # 2 (comment 2.12): "Global mean χ^2 improves by 19% with SEOM-IAS cross-section and by 7% with HITRAN 2016 with respect to HITRAN 2008."

Figure R1 below shows the ratio of χ^2 of the retrieval with HITRAN 2008 and HITRAN 2016 (left) and HITRAN 2008 and SEOM-IAS (right), for one year of data averaged into daily 1° x 1° grid, which shows that SEOM-IAS cross section results in a significantly better χ^2 with respect to HITRAN 2008 and HITRAN 2016. In the sensitivity tests, the only parameter that changed in the retrieval was the spectroscopic database, so any difference in the retrieval results could be attributed to the different spectroscopy. From this we concluded (page 8, line 6) "In view of the better spectral fitting results in the retrieved XCH_4 we have decided to use the SEOM-IAS spectroscopy database".

Regarding the results shown in Table 2, it shows that each of the spectroscopic databases introduces an overall bias that cannot be used as an independent argument to favour a specific

database, as the comparison to GOSAT and TCCON might also be biased because of the specific spectroscopy used in their retrievals. The variation in the scatter of 1 to 3 ppb is not conclusive, as this is negligible if compared to the magnitude of other sensitivities and errors in the retrieval.

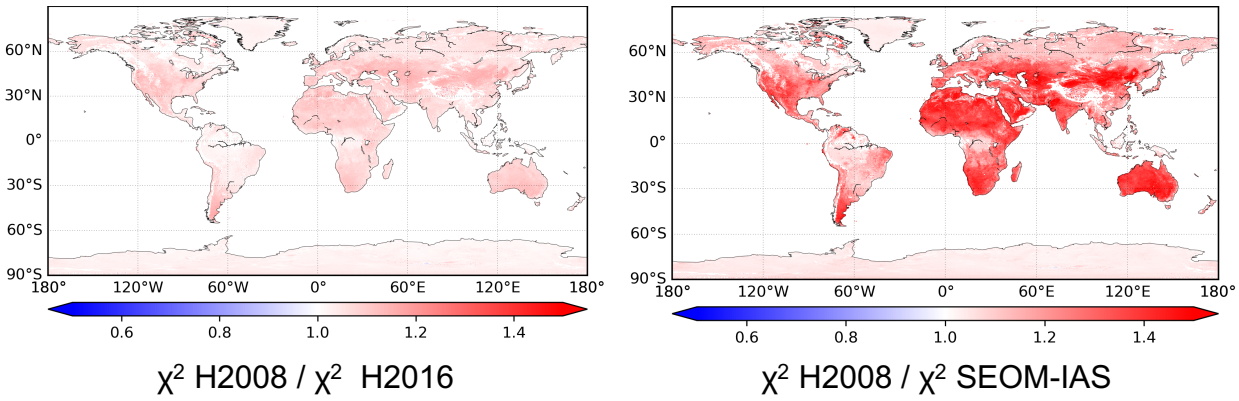

Figure R1: Ratio of χ^2 from the retrieval with HITRAN 2008 and HITRAN 2016 (left) and SEOM-IAS (right).

Comment C 1.13 — Section 3.4. Is this bias correction for albedo really new? As far as I know, the current operational XCH_4 product already offers a bias corrected product. Please clarify.

Reply: Indeed, the operational XCH_4 product already has a posteriori correction applied to it. The novelty of the bias correction presented in this study is the way we have derived it, as we have not used any external or reference data (like GOSAT or TCCON) to estimate the dependence, and the fit to the dependence on surface albedo is done differently.

The new approach is explained in page 10, line 10 – page 11, line 3. Also in page 11, line 19 we refer to the approach in the operational compared to the new fit: "for which the B-spline fit corrects more strongly than the regular polynomial fit that was previously used."

We try to make it clearer by modifying the text:

- Page 10, line 10: "In the baseline operational algorithm few months after TROPOMI was operational, we applied a correction..."

- Page 10, line 12: "[..] we have sufficient data to derive a new ~~the~~ correction".

Comment C 1.14 — Page 12, line 3: surface albedo "A_s": Is this the SWIR albedo? How is the NIR albedo considered?

Reply: In the correction we only consider the surface albedo in the SWIR spectral range, as the dependence of the bias on the surface albedo in the NIR spectral range (see Fig. R2) is negligible compared to the dependence shown in Fig. 3a for the surface albedo in the SWIR.

For clarification, we specify after Eq. 6 that A_s refers to the surface albedo in the SWIR, and in page 10, line 2: "The comparison of TROPOMI [...] shows a dependence of the bias [...] on surface albedo retrieved in the SWIR spectral range".

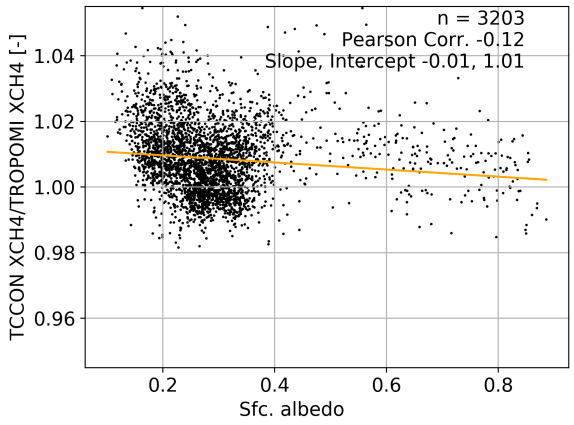

Figure R2: Ratio of XCH_4 measurements by TCCON and TROPOMI as a function of retrieved surface albedo in the NIR spectral range, to compare with Fig. 3a in the manuscript.

Comment C 1.15 — Tab. 3: Add explanation for numbers in brackets. Is this 1-sigma uncertainty?

Reply: The number in parenthesis are the percentage number. We have added to the caption of Table 3: "The table shows [...] (in ppb and in percentages between parenthesis)."

Comment C 1.16 — Typo in CH4 in several places.

Reply: Thank you for spotting this. Changed CH4 to CH_4

References

Barré, J., Aben, I., Agustí-Panareda, A., Balsamo, G., Bousserez, N., Dueben, P., Engelen, R., Inness, A., Lorente, A., McNorton, J., Peuch, V.-H., Radnoti, G., and Ribas, R.: Systematic detection of local CH4 emissions anomalies combining satellite measurements and high-resolution forecasts, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-550, in review, 2020.