Dear Dr. Hu, thank you for your review of our manuscript, and the comments below. In order to respond to your comments, we have kept your original comments in black non-italics. Our responses are in bold blue italics, and changes to the manuscript are in bold blue underlined italics.

General comments: This paper explores the possibility to retrieve the ratio between the two main methane isotopologues from a future GOSAT-2 instrument. In itself, this is a very interesting study, because, if possible, satellite data could be used in the future to discriminate between natural and anthropogenic sources of methane on a global scale. However, the results and conclusions of this study depend heavily on many assumptions, some of which are not quantitatively investigated.

Thank you for raising these points, we address them below.

Main points:

- Impact of precision of current methane retrievals, authors assume here 6ppbv from Yoshida et al. (2011) which is an average and does not include forward model or instrument errors. However, an estimate of the methane precision from TCCON validation is around 15 ppbv (see e.g. Schepers et al. 2012 and Parker et al. 2015). This impact should be quantified or at least discussed.

Thank you for raising this point. We have now expanded a discussion on how the precision of the CH₄ measurements will impact the required ¹³CH₄ precision. The exact details of which are shown in the specific comments section below. However to summarise, the ¹³CH₄ precision is calculated from the range of ¹³CH₄ values that fall into a 10‰ δ^{13} C change, for a given ¹²CH₄ value. The precision of ¹²CH₄ is important as this limits the range of ¹³CH₄ values available for the δ^{13} C change.

In general however, we do not assume that precision errors will be as large as 15 ppbv, since both of these papers use spatial matchup criteria of +/-5 deg when compared against TCCON.

- The authors experiment with different a priori covariance matrices, because no suitable one is known. It seems in this case Philips-Tikhonov regularization is more suitable than optimal estimation. Could the authors comment on that? *Philips-Tikhonov regularization would certainty be suitable in the case of retrieval methane isotopologues. However in the case of GOSAT-2, it is expected that the numerous algorithms currently applied to GOSAT which use a priori covariance matrices (e.g. Parker et al., (2011;2015), Yoshida et al., (2011;2013)) will be applied to GOSAT-2 (via appropriate modification). This paper is aimed at those algorithms, and aims to provide the basic set-up required. We will consider a Philips-Tikhonov method for future follow up studies, and we have included the Philips-Tikhonov method as a discussion point in this manuscript.*

Please see section 9, Page 27, lines 5-13.

- The error analysis could be extended by perturbing the a priori 13CH4 profile with the assumed a priori errors and comparing the retrieved 13CH4 against the truth. Would this be the same as the derived precisions?

This would be an interesting activity, unfortunately at present the author does not have access to an iterative retrieval algorithm (only a linear information content analysis was applied), which we believe would be required in order perform the analysis that is suggested, otherwise we would be passing linear values backwards and forwards, and we are not sure if this would be beneficial to the study.

Specific comments:

- Abstract, page 1, line 22: Rephrase the following sentence for clarity: "Large unconstrained 'a priori' covariance matrices are required ... retrieval errors." Suggestion: "We find that large unconstrained covariance matrices are required in order to achieve sufficient information content, while the solar inclination angle has limited impact on the information content." The authors should avoid "retrieval errors" in this sentence, because that could suggest the solar zenith angle does not have an impact on the forward model retrieval error which is certainly incorrect. *Thank you, we have modified this sentence as requested. We have also changed solar inclination angle to solar zenith angle, as requested by reviewer 2.*

Changed, Page 1, lines 22-23.

- Introduction, page 2, line 19: This sentence seems incorrect: "Plant based photosynthesis enzymes discriminate against carbon dioxide during uptake..." Should it be: "Plant based photosynthesis enzymes discriminate against 13C during carbon dioxide uptake..."?

Thank you for spotting this. We have changed this sentence.

Sentence changed, Page 2, line 19 to "discriminate against ¹³C carbon dioxide (¹³CO₂)"

- In general, the introduction should not contain formulas and derivations. I suggest to move Eq. (1) to a subsection where the requirements on the errors are derived. At the same time, the derivation of the minimum precision of 0.25 ppbv on the 13CH4 retrievals should be made explicitly (e.g. assumed values, error propagation), since this is of such importance to the rest of the paper. In particular, the assumption of a precision of 6ppbv for methane retrievals from Yoshida et al. (2011) is too optimistic, since that is an average and errors up to 15 ppbv can occur. Also, the systematic error is not included in his work or in Yoshida et al. (2011) as stated in the paper, at least a discussion about the impact of the systematic error on the conclusion/results should be given.

Thank you for these observations. We have moved Eq. (1) to a new subsection in section 2, including appropriate aspects of the introduction to give an

appropriate discussion to Eq. (1). This new subsection 2.1 also discusses how the required ${}^{13}CH_4$ precision is derived. The precision is derived by determining what the change in ${}^{13}CH_4$ concentration is for a 10 per mil change in ${}^{13}C$. This is calculated for a range of ${}^{12}CH_4$ values (which are presented in 5 ppb steps), and is represented in the Figure below.



Figure 1. Range of expected terrestrial ¹³CH₄ values (y-axis) given a range of ¹²CH₄ values between 1770 and 1830 ppb, and δ^{13} C between -80 and -10 per mil (x-axis). The diagonal solid lines represent the ¹²CH₄ values for a given ¹²CH₄ value, while varying the δ^{13} C range. There are 13 ¹²CH₄ lines representing the ¹²CH₄ range in 5 ppb steps. The red line (a) shows the ¹³CH₄ change between -50 and -40 δ^{13} C for a ¹²CH₄ of 1770 ppb; (b) is as (a), but includes a ¹²CH₄ change of 5 ppb; (c) is as (a) and (b) but includes a ¹²CH₄ change of 15 ppb.

Based on this figure, we agreed with your statement that we were too optimistic revised down the target ¹³CH₄ precision to 0.2 ppb. This figure does not include precision errors on CH₄. Using this figure, where each black diagonal line represents a CH₄ step change of 5 ppb, we determined that a 5 ppb uncertainty in CH₄ corresponds to a required 0.08 ppb increase in precision of ¹³CH₄ (or 4 per mil δ^{13} C), and a 15 ppb methane uncertainty corresponds to a required 0.16 ppb increase in precision of ¹³CH₄ (or 8 per mil δ^{13} C). We have updated the conclusions as necessary. We have also included a discussion on how an assumed 5 ppb bias on GOSAT methane measurements against TCCON corresponds to a δ^{13} C bias of 4 per mil.

<u>Changes to the introduction w.r.t. moving Eq.(1) to a new section are made on</u> <u>Page 2, lines 28-31; Page 3, lines 9-10, 15.</u>

In regards to the changes described above, Section 2 has been renamed, Page 4, line 11, and the description of the section has been appropriately modified, Page 4, line 12. The new section 2.1, which includes Eq. (1), along with the description of the equation, and the derivation of target ¹³CH₄ precision has been inserted at Page 4, lines 15-30, and Page 5, lines 1-20 and Page 6, lines 1-15.

Based on these changed, the original subsection 2.1 has been updated to subsection 2.2, Page 6, line 16, and the original subsection 2.2 has been updated to subsection 2.3, Page 7, line 13.

- Introduction, page 3, 7: Rephrase/shorten long sentence: "Some measurements from balloon soundings ...to the scientific community" *We have split this into two sentences.*

See Page 3, lines 9 and 10.

- page 4, section 2.1: Mention here that the RTM does not include scattering. *This has been included.*

See Page 6, lines 32-33, and Page 7, lines 1-2.

- page 8, section 4.1: First the authors attempt to determine the variance in 13CH4 by taking the maximum range of observed δ 13C, this approach is a rough approximation at best. From that they derive (3%)2, but nevertheless take (10%)2-(100%)2 in their study. This seems random. Please reformulate or justify better why (10%)2-(100%)2 is reasonable. Also, it not explained in which cases a diagonal covariance matrix is more likely and in which cases an off-diagonal one.

Thank you for raising this point, which is an important point and raised by reviewer 2 as well. We accept that the $(3 \%)^2$ figure is a rough approach, and discuss as such in the updated text.

We have now included a more in-depth discussion into why the $(10\%)^2 \cdot (100\%)^2$ values are used. The reason for this is based on the relationship between the a priori covariance and the DOFS from the assumed GOSAT retrieval. From experience we know that methane covariance is often set to $(10\%)^2$ variance, in order to allow for some variation in the retrieved solution. At this level of variance we can expect between 1 and 2 DOFS (depending on the surface and solar zenith angle). Given that ¹³CH₄ is roughly 1.1 % of the total methane signal, we deemed it very unlikely that setting a $(10\%)^2$ variance for ¹³CH₄ would yield any total column information. We therefore decided to increase the magnitude of variance in order to establish the point when DOFS>1 can be achieved. We accept that such a method will drastically increase a priori and a posteriori errors, but we aim to reduce these through long term averaging.

We have included a discussion in the manuscript to this effect, Page.10, lines 26-31 and Page 11, lines 1-6.

W.r.t to off-diagonal elements, a discussion on why they are necessary has been included.

Please see Page 11, lines 22-27.

- page 12/13, Figure 2 and 3: The color plots are not clear, please use other color scale or representation.

Could you please elaborate on why the figures are not clear? Reviewer 2 has identified all figures as being clear. Thank you.

- page 17/18: It is mentioned that the combined band 2 and band 3 retrieval significantly increases computation time compared to band 2 or band 3 retrieval. Stating the CPU time for all cases would be useful to make that point. *The code/analysis method used in this paper was not an optimized retrieval*

The code/analysis method used in this paper was not an optimized retrieval code, but a linear analysis of the Averaging Kernels and a posteriori errors, incorporating the ORFM, and as such we do not feel that stating the exact CPU time for each analysis would be beneficial, since this will not relate to any fully optimized algorithm. However we have included a rough estimate of the time difference between considering each band individually, and combining them. This number is caveated with the facts stated above.

Please see Page 21, lines 26-29.

- I am missing a discussion on how methane isotolopogue retrievals, if successful, could be validated. Please include a discussion on possible validation strategies, e.g. using NOAA measurements.

Thank you for this important point, we have now included a discussion on potential future validation strategies, including NOAA, CTMs and TCCON.

Please see section 8, Page 26, lines 6-18.

Technical corrections:

-"plant-based" or" plant based", use one consistently throughout the text *Thank you, we have changed this, and similar examples throughout the text.*

page 7, line 13: definite -> define
Thank you, we have changed this.

<u>Page 9, line 25</u>

- page 11: severally - > severely Thank you, this sentence was removed, rephrased and placed in section 2.2 in accordance with the recommendation of reviewer 2.

Please see Page 7, lines 1-3.

Other changes not specified in the above comments:

With inclusion of a new Figure 1 and 5, all of the old figures have been renamed as appropriate, along with all references to the original figures.

In response to a query from reviewer 2, we have inserted an example optical depth plot for ¹³CH₄, in order to emphasize the limited optical depth of ¹³CH₄. *Please see Figure 5 Page 19*,

Based on the modification of the target ¹³CH₄ precision we have updated the results and conclusions shown in Section 7.1, 7.2 and 7.3, revising the target precisions, and the length of averaging times required. Please see, Page 24, lines 9-21, 28-33, Page 25, lines 1-6, 13-14.

Based on the inclusion of discussion points on the Philips-Tikhonov method, and future validation methods, the conclusions and summary section is now section 10.

Based on updates to the precision estimates, we have updates the metrics stated in the conclusions and summary section. Please see Page 27, lines 15-29 and Page 28, lines 1-6.

As above, we have updated all results stated in the abstract.