We thank the reviewer for constructive and helpful suggestions well in advance, and thus allowing us to provide some early responses, which may help us to get further views from the reviewer before the end of the discussion. We appreciate very much this opportunity to discuss online.

Here we would like to share the main directions for improving the article:

- The manuscript structure was revised as suggested
- The content was more adjusted to the AMT requirements
- All requested figures have been replotted

The reviewer's specific comments (shown in blue) are addressed below.

### **Reply to Anonymous Referee #2 comments**

## **General Comments:**

This manuscript presents the vertical distribution of CH4 from GOSAT retrievals over India within the context of elucidating: a) issues related to GOSAT sensitivities and 'a priori' profiles, b) processes influencing the spatiotemporal distribution (emissions, transport), and c) variability across the region. All these aspects are relevant to atmospheric CH4 investigations especially that there are only few retrievals (and less in-situ datasets) available. These three aspects are also described in the paper within a fairly reasonable depth. However, I have two major concerns, which require attention from the authors. That is,

1) The relevance of this study to the scope of AMT is unclear. Unless the paper is refocused on issues with GOSAT retrievals esp the choice of a priori and/or highlighting the sensitivities of GOSAT and the proper use and interpretation of these retrievals. The paper already presented several figures and discussion to these points but more emphasis could be made to bring it closer to the scope of AMT.

We agree the topic of this study is quite broad due to the complexity of the research topic and several data sets used in the analysis. To meet the requirements of AMT, we decided to narrow the range of studies to GOSAT-TIR specific subjects mainly.

2) Lacks comparison (verification) with available independent measurements. While it is understandable that there are only few measurements available, model-based comparisons are not sufficient. Some efforts to compare with other measurements (aircraft or ground based or other retrievals from different instrument) would strengthen the paper's findings.

As shown above, this issue is described in Discussion L.416-426: "Despite essential progress, the development of satellite methods for studying atmospheric methane is obstructed by a number of limitations. The launch rate of new orbital instruments is significantly ahead of the development of

a ground-based and aircraft measurement network for their validation. Due to the scarcity of suitable aircraft observations over India, validation of GOSAT-TIR profiles cannot cover a variety of seasons and land regions studied in this work. However, in the newly prepared paper by N. Saitoh "Intensive validation analysis of GOSAT/TANSO-FTS thermal infrared CH<sub>4</sub> data (version 1) based on aircraft observations" (to be submitted to "Remote Sensing"), the intensive validation work of GOSAT-TIR CH<sub>4</sub> profiles is described. In this paper, global comparisons are conducted based on HIPPO, CARIBIC, JMA, and CONTRAIL/ASE aircraft observations. In low latitudes corresponding to the India location, compared datasets include CARIBIC profiles over MAA (Chennai, India), BOG (El Dorado, Colombia), and CCS (Venezuela) airports and CONTRAIL/ASE over GUAM (US) airport. The validations show that TIR V1 CH<sub>4</sub> and aircraft CH<sub>4</sub> profiles agreed with each other within 10-15 ppb and there was no evident seasonal dependence in the CH<sub>4</sub> differences."

#### **Specific Comments:**

## 1) Title is a bit misleading as the paper does not discuss this in depth.

We agree. The title was revised as "Interpretation of GOSAT CH4 vertical profiles over the Indian subcontinent: effect of a priori and averaging kernels".

2) Abstract states that the objective is to understand retrieval sensitivity, but the results are more towards comparison of CH4 variations across with models including emissions, without any independent measurements to compare with.

The text of the paper was seriously elaborated to meet this requirement.

3) Line 16: Stating "22 vertical levels ... provide critical information' is misleading. Might be better to state its DOFS and vertical sensitivities.

Agree. Revised as follows L.15-17: "A comparison of modeled and retrieved CH4 vertical profiles shows the GOSAT/TANSO-FTS TIR sensitivity is sufficient to provide critical information about transport from the top of the boundary layer to the upper troposphere and lower stratosphere in a consistent manner."

## 4) Line 18: 'excepting' ?

Revised as L.18-19: "...50 ppb, except for the altitude range above 150 hPa, where..."

5) Line 95-100. It would be great to describe the retrieval algorithm including a priori error covariance assumptions (if this is an optimal estimation). A short description as well of NEIS relative to MIROC (esp emissions used in NEIS).

The retrieval algorithm description was updated L.96-99: "The retrieval algorithm for the TANSO-FTS TIR V1 CH<sub>4</sub> product is basically the same as for the V1 CO<sub>2</sub> described in Saitoh et al. (2016). It adopted a nonlinear maximum a posteriori (MAP) method with linear mapping. *A priori* covariance matrix for CH<sub>4</sub> in the V1 CH<sub>4</sub> retrieval is set to be a diagonal matrix with vertically fixed diagonal elements with a standard deviation of 4%."

NIES simulation setup was updated L.100-106: "For simulation NIES TM used the monthly varying flux for 2000 (575 Tg yr-1) based on the Emission Database for Global Atmospheric Research (EDGAR) version 32FT2000 (Olivier and Berdowski, 2001) for anthropogenic CH4, and on GISS emissions (Fung et al., 1991) for natural CH4, as obtained from Patra et al. (2009). The chemical destruction of CH4 by OH radicals was calculated based on climatological monthly mean OH radical concentrations (Spivakovsky et al., 2000) and a temperature-dependent rate constant."

# 6) Line 105-110. This is a very useful discussion of GOSAT retrievals. Why are these other retrievals not used for comparison over India in this study?

We think "these other retrievals" mean other CH<sub>4</sub> data from AIRS, ACE-FTS, and MIPAS. ACE-FTS and MIPAS are solar-occultation and limb-viewing sensors, respectively, so their horizontal resolutions are too law and their measurements are too sparse to discuss detailed features of CH<sub>4</sub> over India. AIRS has a much lower spectral resolution than GOSAT, so GOSAT is more suitable for the discussions of CH<sub>4</sub> vertical distributions over India.

7) Section 3.2. This is also a very useful section. If DOFS is 1, why do we have profile information? DOF means the number of purely (mathematically) independent pieces of information. The real atmospheric layers correlate with each other, so even if its DOF is one, TIR has some sensitivity to lower tropospheric CH4 concentration judging from small differences seen between TIR and a priori CH4 concentrations in the lower troposphere. However, the sensitivity to the lower troposphere is smaller than the upper levels and we should not treat TIR CH4 data in the lower troposphere in a similar manner to those in the upper levels; therefore, we more focus on the upper troposphere where TIR measurements has the most sensitivity in the revised manuscript.

Added to the text L.135-138:" The degrees of freedom (DOF) of signal for CH<sub>4</sub> observation by GOSAT-TIR band (V1 algorithm version) is around 1 over low-latitude part of India. DOF means the number of purely (mathematically) independent piece of information. However, the real atmospheric layers correlate with each other, so even if its DOF is close to 1, TIR has ability to derive new knowledge about CH<sub>4</sub> concentration."

8) Line 213. 'resampled' ? 'resampled' -> regridded



9) Line 221. It may be interesting to show differences in AK over land and ocean.

Updated fig 1 include AK for two land and one oceanic regions as show above.

10) Figure 4 & 5. These figures are informative.

Agree. Thank you

11) Figure 9, Line 284-287. What about the retrieval errors (from the a posteriori estimates)? Please elaborate 'we found that differences between a priori and retrieved CH4 profiles are larger than its retrieval error...'.

This sentence revised L.309-314: "The variation of GOSAT-TIR sensitivity are taking into account by the implementation of the *a priori* profiles and AK functions (Fig. 1) to the modelled data sets (ACTM<sub>Cao</sub><sup>AK</sup> and ACTM<sub>WH</sub><sup>AK</sup>). The variabilities shown in Figure 10 are larger than GOSAT-TIR retrieval errors, derived here as the diagonal elements of the posteriori error covariance matrices based on the MAP method, which include random error components of the retrieval. Therefore, GOSAT-TIR and model show good agreements (mismatch is inside 1- $\sigma$  STD uncertainty) within both errors (natural variabilities and retrieval random errors)."

#### 12) Line 311-312. This sentence is unclear. Please restate.

This sentence revised L.342-346: "Fig. 11 shows the use of GOSAT-TIR AK functions have significant smoothing effect, approaching the MIROC4-ACTM model profiles to *a priori* so much that the difference between the calculations for the Cao and WH emission scenarios becomes barely distinguishable. This is especially visible above the level of 150 hPa, where the sensitivity of GOSAT-TIR there drops sharply and the satellite retrievals and the AK convolved model profiles strongly follow the *a priori* profiles.

"

13) Line 316-317. Is this a study where different a priori profiles (and I assumed the error covariance is the same) are used in the retrievals. Please make sure the use of 'a priori' is consistent across the manuscript (including italics and non-italics).

Revised through the text

## 14) Line 320-324. This is a useful discussion and should be highlighted more.

Discussion of *a priori* profiles was extended L.432-456:

"A priori profiles play an essential role in processing a satellite signal, especially for the CH<sub>4</sub>, which has a significant change in a lifetime with altitude. The choice of such a profile (usually provided by model calculations) is a critical point since due to small DOF the retrieval algorism cannot overcome large errors in input data. The TransCom-CH<sub>4</sub> experiment [Patra et al., 2011] showed a significant scatter between the participated models, including the NIES model later selected for calculating GOSAT-TIR *a priori* profile (described in 2.2). In this study, the difference in the methane profile gradient, its seasonal variability (winter and summer) between *a priori* and the MIROC4-ACTM model was revealed in the UTLS zone (levels of 150-20 hPa). Apparently, the difference in the modelling of UTLS is a key factor, as the MIROC4-ACTM meteorological parameter is driven by recently updated reanalysis and its vertical resolution (67 sigma-pressure levels) is quite higher than that of the NIES (47 sigma levels). Even more important, the stratospheric part of the NIES model was adjusted to the observed age of air for CO2 and long-term satellite observations from HALOE for CH<sub>4</sub> [Saeki et al. 2013]. Therefore, the reason for the misfits (GOSAT-TIR *a priori* vs ACTM) extremely controversial without additional studies with the use of custom *a priori* profiles in retrieval.

In the case of long-term projects, the updating of *a priori* data in accordance with the current progress becomes important. From the moment of the GOSAT launch, the calculation of *a prior* profiles is carried out according to the same scheme. This is important for the long-term consistency of the GOSAT-TIR CH<sub>4</sub> product but does not take into account the recent improvements (e.g. new reanalysis data, higher vertical resolution, and convective parameterizations) implemented in MIROC4-ACTM [Patra et al., 2018], as well as further understanding of the CH<sub>4</sub> budget [Saunois et al., 2020].

As the release of a new version of retrieval algorithms designed to improve and update satellite products is not regular, perhaps, for further progress, the retrieval process should be open access with a possibility of use of custom *a priori* information. Therefore, this will require greater transparency of technical information from satellite projects and significant optimization of retrieval calculations, since such tasks are requiring large computational resources. Promising is performing of retrieval inter-comparison projects (one algorithm with a variety of *a priori* information and set of algorithms with the same of *a priori*), as it was done for CTMs development [Patra et al., 2011]. However, this *a priori* profile issue remains hidden (but no less relevant) in the field of the main efforts of the scientific community working with the column-averaged burden (XCH<sub>4</sub>) derived from the SWIR band."

## 15) Figure 11. More discussion on this (relative to a priori) would strengthen this paper.

The following sentences are added to L.361-366: "Apparently, the *a priori* profiles from NIES model shows insufficient vertical transport due to incomplete convective parameterization required to simulate tracer transport under monsoon conditions (Fig. 12). This problem forced the transition to a more sophisticated reanalysis JRA-25/JCDAS (Japanese 25-yr Reanalysis/Climate Data

Assimilation System developed by the Japan Meteorological Agency (JMA)) and the adaptation of the new parameterization as described by Belikov et al. [2013]. The GOSAT-TIR retrieval is trying to compensate for such a concentration deficit in the upper troposphere. On the other hand, the overestimation by MIROC4-ACTM despite the implemented modifications [Patra et al., 2018] is also possible."

16) Section 3.5. This looks like more of a comparison with ACTM and elucidating differences. It may be better if this can be made a separate section with slightly different heading.

Section 3.5 was spited into 2: 3.5 Seasonal variation of CH<sub>4</sub> and 3.6 Regional CH4 emission estimation

# 17) Line 375. 'ACTM WH is superior to ACTM CAO'. It's unclear from the bar graphs.

This sentence is related to new Fig.13 now, where ACTM<sub>WH</sub> (blue line) is higher than ACTM<sub>Cao</sub> (red line) for all 3 levels (panels a-c). For more details, the simulation difference (ACTM<sub>WH</sub> - ACTM<sub>Cao</sub>) is shown by dashed line.

#### 18) Line 399. What is the basis for 10-15 Tg yr overestimation (How was this number derived?)

Revised text added to L.469-478: "The problem of reliable data lack for estimation regional CH<sub>4</sub> budgets can be mitigated by GOSAT-TIR CH<sub>4</sub>. Relying on our comparison (Fig. 14), we suggest the Cao flux combination with the annual mean emission of 65.7  $\pm$  5.8 Tg yr<sup>-1</sup> for the period 2009-2014 as more plausible. This confirms the assessment made by Patra et al. [2016], indicating that the EDGAR inventory (version 4.2FT2010) with a value of 73.3–83.2 Tg yr<sup>-1</sup> overestimated the South Asia regional emission by 10-15 Tg yr<sup>-1</sup>. A significant part of the extra fluxes is concentrated in a few relatively small regions in the Northen India (fig. 2). However, our best estimate emission of 51.2  $\pm$  4.6 Tg yr<sup>-1</sup> over India is much greater than 19.6–24.3 Tg yr<sup>-1</sup> estimated by [Ganesan et al., 2017], combined *in situ* data of different time coverage and SWIR CH<sub>4</sub> retrievals in the trajectory-based modelling framework. Simulation with two scenarios showed that during the monsoon significant CH<sub>4</sub> amount due to extra fluxes can fast propagated to the UTLS zone and not been detected by ground-based measurements. This emphasizes the importance of correctly accounting for the effects of vertical transport for emission estimating."

19) Conclusions. While comparison with models is informative, it remains to be proven if the differences between GOSAT and modeled profiles reflect 'real' differences — unless independent measurements (and/or retrieval experiments) are made.

As shown above, this issue is described in Discussion L.416-426.