This version has been updated in accordance with the comments by Anonymous Referee #2 and the final version of the article.

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.
- To avoid misunderstandings, all considered time periods have been fixed to 2009-2014.
- All requested figures have been replotted using a priori data and the averaging kernels.

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

Reply to Anonymous Referee #1 comments

Review of the manuscript "Methane vertical profiles over..." by Belikov et al. submitted to the journal Atmospheric Measurement Techniques.

The article "Methane vertical profiles over..." by Belikov et al. presents the vertical distribution of methane (CH_4) retrieved from the GOSAT TIR measurements focussing on the Indian subcontinent over the period 2009-2014. Coupled to a model (MIROC4), the seasonal variations of CH_4 in the lower, middle and upper troposphere are analysed in terms of transport and sources. The sensitivity of the GOSAT CH_4 retrievals into the lowermost troposphere is discussed in order to quantify a surface emission of CH_4 over India of 51.2 +/- 1.6 Tg yr-1 in 2009-2014.

It is a potentially very interesting paper that may help in the quantification of the CH_4 flux over a key source area, namely India and its surroundings. Nevertheless, the article has too many weaknesses to properly address this fundamental issue. In my opinion, the manuscript has a too broad (not precise and/or not rigorous enough) approach of the scientific and technical issues related to CH_4 in the observation and in the modelling aspects. These aspects will be dealt in detail below but major ones are: 1) a title that is too vague but consistent with the content of the manuscript, in other words the paper is a mixture of observations, validation, modelling and process studies, but none of them are carefully addressed; 2) the lowermost tropospheric sensitivity of the CH_4 observations in the TIR is not satisfactorily addressed/proved since it is too much impacted by the dynamical a priori information used; 3) the 2 model outputs in the lowermost troposphere are compared to observations of GOSAT without using averaging kernels (Fig. 13), consequently no quantitative information can be derived on the CH_4 emissions from this. I would propose to the authors to focus on one important aspect of their study, namely the CH_4 emissions over India, and to clearly show that GOSAT TIR observations can deal with that. Unfortunately, for all these reasons, I cannot suggest to major revise the manuscript but rather to reject it, and to resubmit a version focussing on one single aspect of their analyses.

MAIN POINTS

1) TITLE & CONTENT

The title, very consistent with the content of the study, is too vague to properly address the rationale and the scientific outcomes of the study. Although the geographical extent of the project is clearly presented, the analysis shows CH_4 fields analysed: 1) in three different layers from the lowermost to the uppermost troposphere (800, 500 and 200 hPa), 2) over different time periods: from 2009 to 2014, but focussing on 2011 (with no explanation to why focussing on this particular year) and highlighting climatological fields over 1981-2010 and 2009-2014 underlining the lack of consistency in the data analyses used in the overall study (Figure 2), and 3) combined with 2 model outputs that are not clearly presented in terms of differences of processes impacting CH_4 emissions (section 3.1). This wide range of studies are not rigorously addressed and, by the end of the article, the conclusions are not supported by the presented analyses.

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. As a result, the title was reformulated as "Interpretation of GOSAT CH4 vertical profiles over the Indian subcontinent: effect of a priori and averaging kernels".

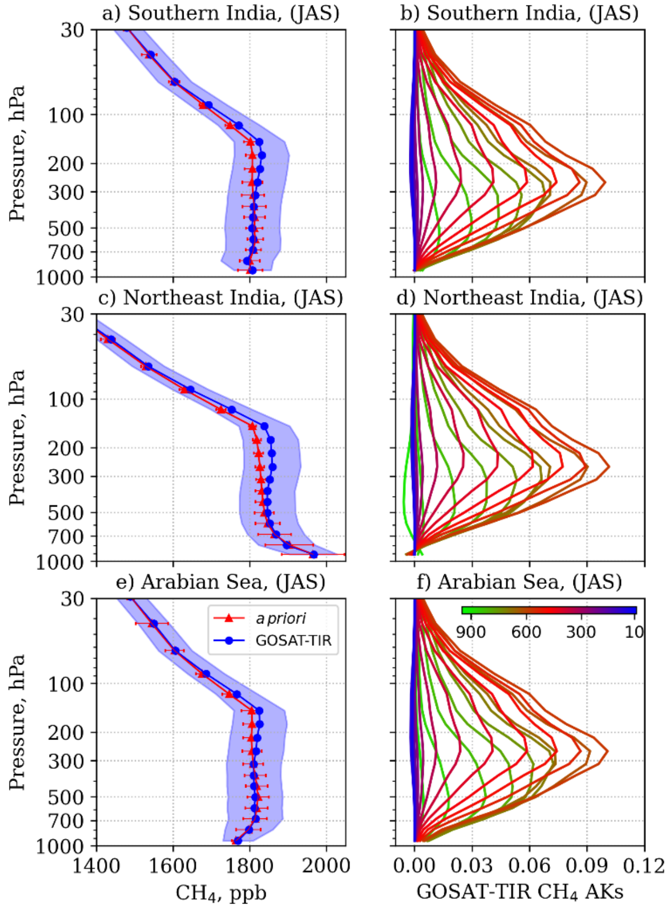
As stated in the manuscript "This study uses the GOSAT-TIR CH_4 product (version V1), which is released for the period from April 23, 2009, through May 24, 2014". We also focused on 2011 to plot seasonal distributions of specific parameters on lat-lon grid (Fig. 5-9), as this year has fewer gaps in GOSAT-TIR observations. For meteorological fields depicted in Fig. 4 climatological values were replaced with ones for 2011. To address time variability time series and time-altitude cross-sections were used (Fig. 12, 14).

The used two model outputs are related to CH_4 emissions from wetland as described in two different schemes developed by Cao et al. [1996] and Walter et al. [2001]. The main difference between these approaches in most cases can be explained by the combined effect of changes in soil temperature and the position of the water table. The dependence of CH_4 emissions on water table depth is complicated and highly non-linear, so the strength of the fluxes is mostly variable in lowland areas along large rivers. In general, the WH scheme fluxes are about 5-10% larger the Cao, excepting the WIGP, EIGP, and NEI regions of India and Bangladesh where the maximum difference reaches 20-40% (Fig. 2). Besides, there are small hot spots in Southeast Asia (e.g. Mekong River Delta).

2) LOWERMOST TROPOSPHERE

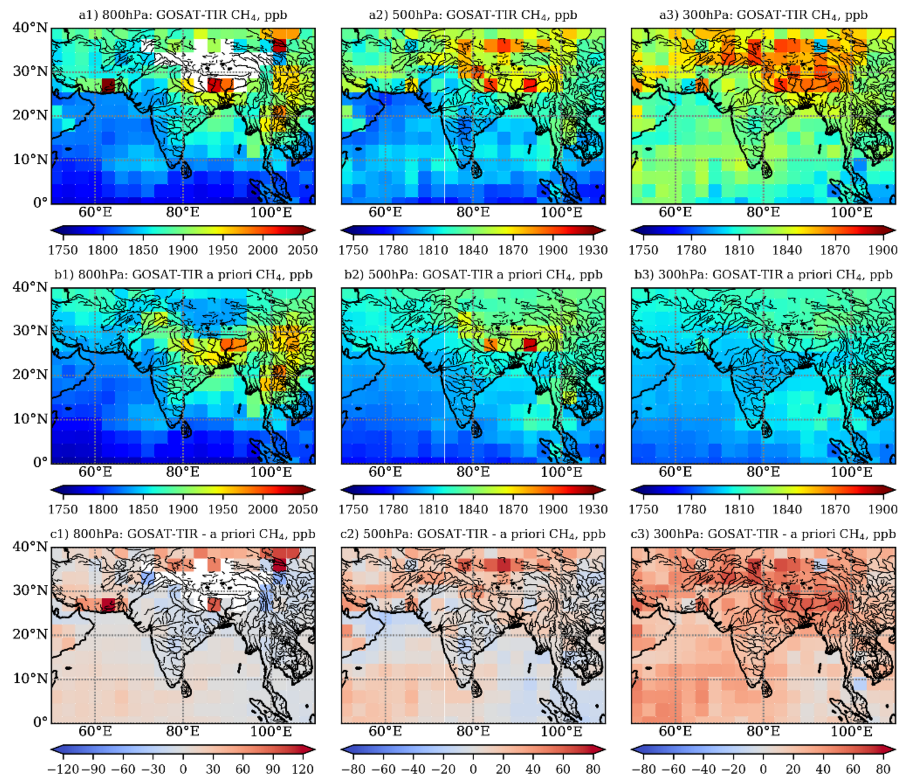
The analyses of CH_4 in the lowermost troposphere are probably the most interesting results presented in the manuscript. Unfortunately, the authors fail to convincingly show that the GOSAT TIR CH_4 observations are actually sensitive to 800 hPa. One of the main reasons is the use of an a priori information that is issued from a model, that is to say, that is dynamically evolving in time and space. As a consequence, the CH_4 retrievals (whatever the layers considered) are contaminated by this dynamical a priori. Figure 3 left shows typically the obvious relationship between retrieved CH_4 and a priori CH_4 around 800 hPa (no differences) that is also shown in Figure 3 right where the averaging kernels are peaking at 300 hPa and, for few of them, very difficult to examine since they are labelled in levels and not in pressure, around 500 hPa. Maps of CH_4 in the lowermost troposphere at 800 hPa (Figures 4 and 5) also show the strong a priori contamination to the GOSAT CH_4 observations over India with almost similar fields in GOSAT and in the a priori CH_4, that is not the case at 500 and 200 hPa. The vertical distribution of CH_4 (Figures 9 and 10) also highlights this point between GOSAT, a priori and the 2 model outputs at 800 hPa. I would suggest to particularly focus on this issue and carefully show to the reader that GOSAT TIR can actually observe in this layer.

For easy examination, labels of figure 1 (right panels) was updated to show pressure rather than levels.



New figure 3 (Now figure 1): Seasonal mean (July-September 2011) over regions of Southern India (upper panel), Northeast India (middle panel), and Arabian Sea (bottom panel). The left columns show the GOSAT-TIR CH_4 *a priori* with 1-σ STD uncertainty (red line with error bars) and GOSAT-TIR CH_4 profile with the retrieval error (blue line with shaded area); the right columns depict AK matrix of GOSAT-TIR CH_4 retrievals averaged over time. There are 22 lines for GOSAT-TIR retrievals, corresponding to the retrieval layers used in each of them.

Following the recommendations of the reviewer and the requirements of the AMT journal, the main research focused on GOSAT-TIR observations in the middle and upper part of the atmosphere (500-300 hPa), where the sensitivity of the TIR instrument is relatively high. For this the corresponding figures have been updated using a priori data and the averaging kernels.

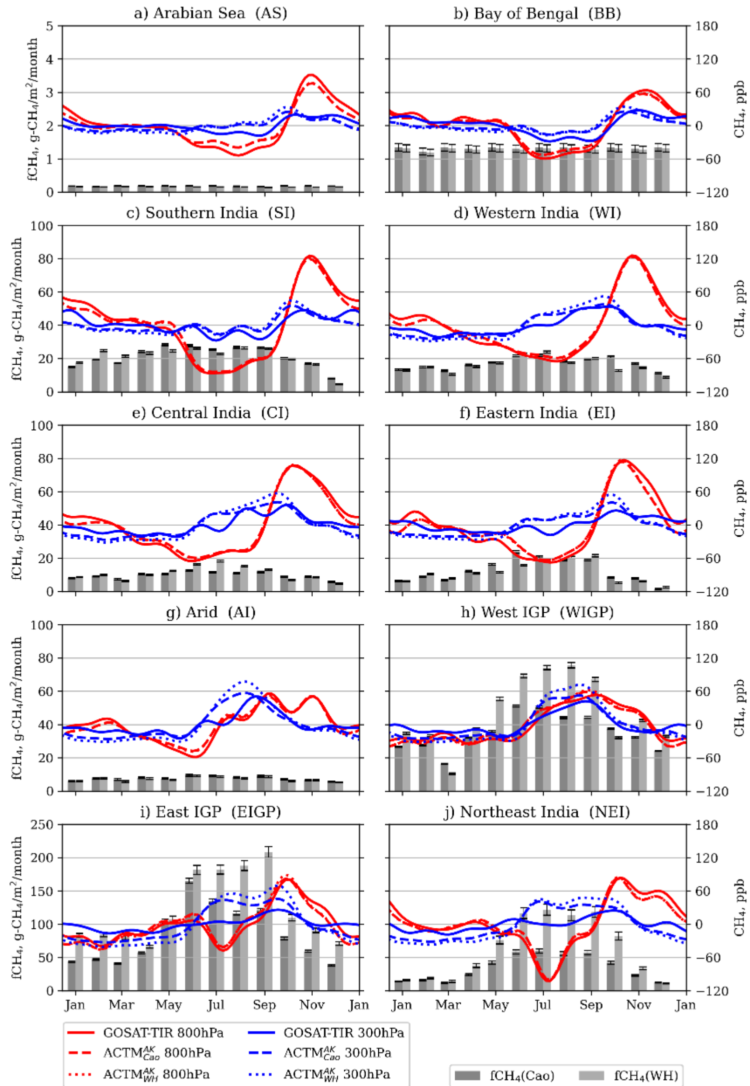


New figure 4 (now figure 6): Latitude-longitude distributions of CH_4 at the levels of 800, 500, and 300hPa (the left, middle, and right panels respectively) observed by GOSAT-TIR for the season AMJ 2011. The first upper panels (a1-a3) show GOSAT-TIR CH_4, the second panels (b1-b3) show GOSAT-TIR *a priori* CH_4, and the third panels (c1-c3) show difference between the GOSAT-TIR observed and *a priori* distributions, respectively.

3) CH_4 EMISSIONS

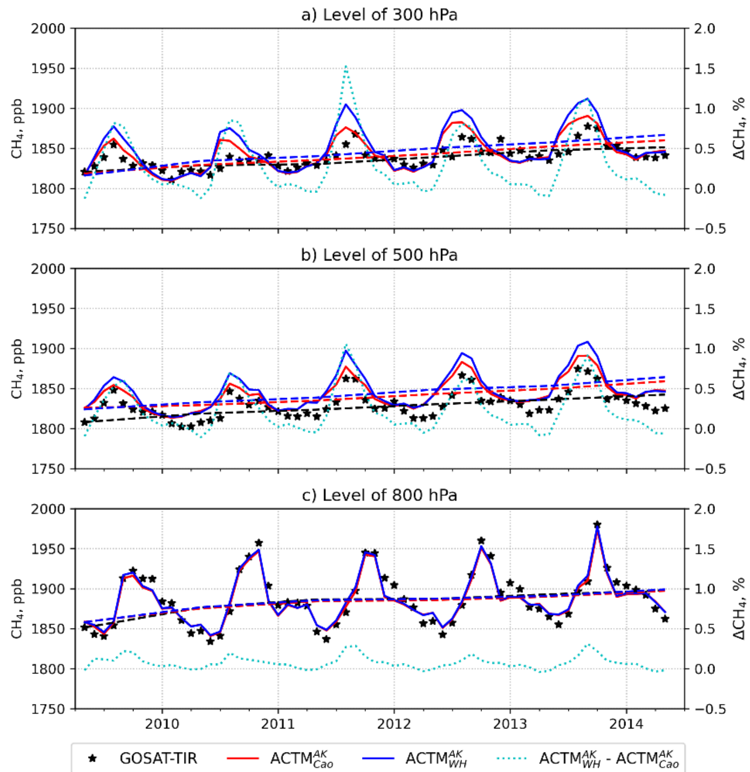
The two CH_4 emissions used in the article will need to be properly presented and explained in what and why they were employed for the study. Differences in the modelled CH_4 fields are clearly shown although some Figures will need to be better presented as for instance Figure 12 that does not present the period over which the data are highlighted. The most important issue in my opinion comes from Figure 13 for which the modelled CH_4 (ACTMCao and ACTMWH) are not convolved with the a priori information using equation (1), that is to say, it is impossible to quantify (as the authors do) the South Asian regional CH_4 emission simply based on this Figure since models and observations are not in same space. A more rigorous comparisons will need to be done in order to infer such an important value.

Figure 12 shows multi-year (2009-2014) seasonal variation of CH_4 from GOSAT-TIR and MIROC4-ACTM. To avoid misunderstanding we updated this information to the caption. The plot was also updated.



New figure 12 (now figure 13): Multi-year (2009-2014) seasonal variation of CH_4 (right y-axis) derived by the implementation of the Prophet model for levels of 800 (red lines) and 300 hPa (blue lines) over considered regions from GOSAT-TIR (solid line), $\mathrm{ACTM}_{\mathrm{Cao}}^{\mathrm{AK}}$ (dashed line), and $\mathrm{ACTM}_{WH}^{\mathrm{AK}}$ (dotted line), respectively. The background bar plots represent Cao (dark grey) and WH (light grey) CH_4 fluxes (left y-axis), respectively. Please note the different scale of y-axes (left) for fluxes.

Figure 13 was replotted using AK and a priori information using equation (1).



New figure 13 (now figure 14): Time series of CH_4 averaged over the area of South Asia for levels of a) 300, b) 500 and c) 800 hPa. Symbols state the GOSAT-TIR observations, red and blue lines are for $\mathrm{ACTM}^{\mathrm{AK}}_{\mathrm{Cao}}$, and $\mathrm{ACTM}^{\mathrm{AK}}_{\mathrm{WH}}$, respectively. Solid and dashed lines are for monthly and yearly averaged concentrations (left y-axis), dotted line shows the difference between the model simulations (right y-axis), respectively.

Minor Points

I will not list in detail all the minor points that will need to be improved but only the key ones for a future version.

1) In general, please quantify (absolute and relative values) when making comparisons (differences for instance) and avoid terms as “good agreement”. This happens several times in the manuscript.

The corresponding sentences were revised.

2) Clearly present which version of the GOSAT CH_4 retrieval you have used, whether this version has been validated or not and, above all, if the validation has also focussed on the lowermost troposphere (800 hPa).

Agree. Due to technical issues in the typeset process, the version mark (V1) disappeared in several places. Thus, the version was added one more time, for example: “This study uses the GOSAT-TIR

CH_4 product (version V1), which is released for the period from April 23, 2009, through May 24, 2014."

This version was validated in several recent works [Holl et al., 2016; Zou et al., 2016; Olsen et al. 2017], as stated in the Introduction. However, the lowermost troposphere (800 hPa) level is not covered in those works.

This issue is also described in Discussion LL.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_4 data (version 1) based on aircraft observations" (to be submitted to "Remote Sensing"), the intensive validation work of GOSAT-TIR CH_4 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_4 and aircraft CH_4 profiles agreed with each other within 10-15 ppb and there was no evident seasonal dependence in the CH_4 differences."

3) Explain whether you have used daytime or night-time or both CH4 retrievals since this may impact on all of your results, particularly in the lowermost troposphere.

The following sentence was added L.127-132: "GOSAT measures in TIR spectra twice a day. The first one is performed along with the SWIR band at local noon ±1 h, the second one near midnight (local noon ±12 hr). Daytime observations are being screened more thoroughly using the Cloud and Aerosol Imager (CIA). At night, filtering is carried out only by the TIR algorithm. Therefore, the number of scenes of daytime and night-time observations can differ significantly, especially during the monsoon period. However, the average difference between daytime and night-time observed CH_4 profiles is usually within ±20 ppb. We leave a more detailed analysis of the daily variation of methane outside the scope of this work."

4) Prophet should be presented in the section "Method" and we should understand why you have used this model. Prior to show the results, you should clearly highlight your methodology to achieve your scientific goal(s).

The Prophet section was moved to "Method":

2.7 The Prophet analysis and forecasting model

GOSAT-TIR CH_4 observations shows large temporal and spatial heterogeneity, which complicate deriving of seasonal cycle variation. To derive cyclic variations from noisy and time irregular datasets use of special methods is highly desirable. The Prophet is a novel time-series analysis and forecasting model [Taylor and Letham, 2018], which performs smoothing of time-series data based on a generalized additive model with three main components: trend, seasonality, and holidays. Compared to traditional exponential smoothing, Prophet can easily handle temporal patterns with multiple periods and has no requirements regarding the regularity of measurement spacing. The model has a robust performance in the presence of missing data and trend shifts and typically handles outliers well while working with time-series that have several seasons of historical data with strong seasonal patterns. The Prophet manage to use of all data points for the study period, thereby increasing accuracy and reducing sensitivity to random outliers [Belikov et al. 2019]. Later in the text:

3.5 Seasonal variation of CH_4

The Prophet time-series analysis and forecasting model [Taylor and Letham, 2018] was implemented to derive mean seasonal cycle of CH4 from GOSAT-TIR and MIROC4-ACTM for the levels of 800 and 300 hPa for 2009-2014 ignoring year-to-year variations (Fig. 13).

5) Why did you use so many periods? If important for you, please explain.

To avoid misunderstandings all considered time periods were revised, as stated above.

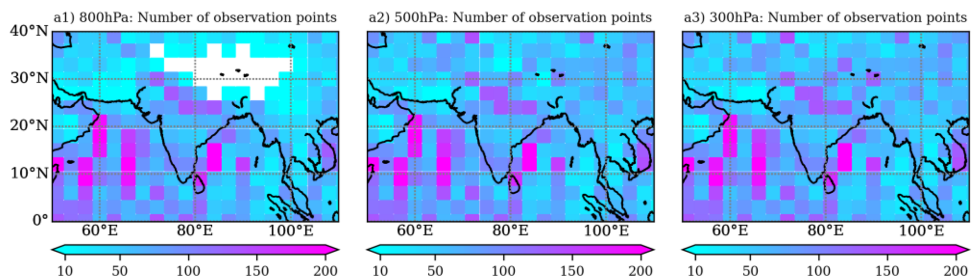
6) The Degree of Freedom (DOF) of the CH_4 TIR retrievals is "around 1" (L. 192), that is to say, you only have access (in theory) to a columnar information of CH_4 from TIR. So, explain how you can still have a sensitivity in the lowermost troposphere with DOF=1.

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_4 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_4 concentration."

7) The colour Table (yellow-red) showing the Number of observation points in Figures 4 and 5 does not really highlight the dynamical range over the Indian subcontinent. I would propose to modify it.

The colour map of the figure was modified.



New figure 4: Latitude-longitude distributions of GOSAT-TIR CH_4 observation points numbers at the levels of 800, 500, and 300hPa (the left, middle, and right panels respectively) for the season AMJ 2011.

8) Some sentences are difficult to understand (English and/or science): L. 204, 243, 257, 260, 356, 366, 413.

We did our best to revise the pointed sentences

9) In general, I would propose a section dedicated to the discussion of the results obtained at least to confront your results to those of other studies.

Agree. The discussion section was added.