

Responses to referee 3: (received: 22 August 2019)

We thank the referee for very insightful questions and comments. They have helped to improve the quality of the paper. We have substantially revised and reorganized the manuscript, in many parts extended paragraphs has added. Our responses are given point-by-point below (blue Times New Roman font) following each of the reviewers' comments, which are repeated in full (black Times New Roman Italic font). Reproduced text from the revised manuscript is set in green Times New Roman font.

In addition to the change made in the manuscript to take into account your comments or the comments of the other referees, several other changes have been made and are listed here.

General Comments

This manuscript presents an investigation of the large uncertainty of MIPAS V5R_CH4_220 methane retrievals in the tropical upper troposphere and lower stratosphere and attributes it to interference from atmospheric water vapour. Including water vapour when fit in methane reduces the uncertainties in the new data version MIPAS V5R_CH4_224.

The work is a useful contribution to the field and appropriate for AMT, but the manuscript needs major revisions and another round of reviews.

I have read the reviews of the other two referees. My assessment of the manuscript is similar to theirs, so I will not repeat their specific and technical comments.

Response: The response to other referees has been responded and some of the response were also added here.

In general, the manuscript should be improved in the following ways:

Response: We would like to thank the reviewer for this positive evaluation. This paper has addressed the cause of large uncertainty in the old version of MIPAS CH₄ in the upper troposphere and lower stratosphere of tropics which is due to water vapour variability. However, the contribution of water vapour in the uncertainty of new data sets of MIPAS CH₄ had been reduced as water profile is jointly retrieved.

Change the title. MIPAS is not a satellite. e.g., "The impact of H₂O variability on the accuracy of MIPAS CH₄ measurements [or retrievals] over the tropics:

Response: As the referee suggest, we will replace the word "satellite" by "measurements" in the title and written as follows: "Impacts of H₂O variability on accuracy of MIPAS CH₄ measurements over the tropics"

- *Rewrite the abstract for clarity, conciseness, and grammar. The explanation of the results is wordy and unclear.*

Response: In the first sentence of the abstract, before the period we added "at upper troposphere and lower stratosphere" to make clear where the uncertainties are large

Page1lines 4-6: has been replaced by “Coincident measurements by MIPAS, ground based FTIR and MLS of CH₄, H₂O are used to estimate the standard uncertainty of MIPAS_CH4_220, MIPAS_CH4_224 and natural variability of H₂O. Moreover, MLS of CH₄ were derived from EOS MLS coincident measurements of atmospheric water vapour (H₂O), carbon monoxide (CO) and nitrous oxide (N₂O).”

Page1lines 6-8: has been replaced by ” Different methods such as bias evaluation differential method and correlation analysis are employed to explore the latitudinal variations of standard uncertainty of MIPAS_CH4_220, MIPAS_CH4_224 and natural variability of water vapour.”

Response: The results in the abstract have been rewritten as follows:

The averaged bias between MIPAS_CH4_220 and ground-based FTIR measurements in the altitude rang 15-22 km are 12.3%, 8.9 % and -1.2 % for tropics, mid-latitudes and high latitudes, respectively. Whereas the averaged bias for MIPAS_CH4_224 is 3.9 %, -2.8 % and -2.4 %. The average estimated uncertainties of MIPAS CH4 220 methane were obtained 5.9 %, 4.8 % and 4.7 % at altitude ranges of 15 to 27 km for tropics, mid-latitudes and high latitudes, respectively. On the other hand, the average estimated uncertainties of MIPAS CH4 224 methane were obtained 2.4 %, 1.4 % and 5.1 %. Moreover, the correlation coefficient between MIPAS CH4 220 and MIPAS V5R_N2O_220 in a global scale of gridding space 30 degree latitude and 3km altitude found that 0.30, 0.98 and 0.96 in the lower stratosphere of tropics, mid and high latitudes respectively. Nevertheless, the correlation coefficient between MIPAS CH4 224 and MIPAS V5R_N2O_224 are 0.62, 0.80 and 0.66.

- *The manuscript needs much clearer explanations of methods and results throughout, and more detail on how results were obtained.*

Response: As the referee suggested on clarity the manuscript in a way that readers can easy understand by adding detail explanation of methods and results.

P3L2-5: the sentences have been replaced by “The coincident measurements Of H₂O, CH₄ and N₂O by MIPAS, ground based FTIR and MLS were used to estimate the uncertainty of MIPAS_CH4_220 and MIPAS_CH4_224 profiles and the natural variability of H₂O. MLS CH₄ was derived from EOS MLS coincident measurements of atmospheric water vapour (H₂O), carbon monoxide (CO) and nitrous oxide (N₂O).”

Inserted after the period in P3L4; Different methods has applied to determine uncertainty of MIPAS_CH4_220, MIPAS_CH4_224 measurements and variability of water vapour at the three latitudinal bands. Intercomparison results of methane (CH₄) measured by MIPAS with the ground based FTIR products obtained from Addis Ababa FTIR observatory and other two NDACC FTIR sites (Jungfraujoch, Switzerland and Ny-Ålesund, Spitsbergen). It has been analyzed using the statistical analysis methods detailed in von Clarmann (2006). Natural variability of water vapour and uncertainties of MIPAS methane can also be determined using differential method proposed by Fioletov et al. (2006) and applied on different literatures (Toohey et al. (2007); Sofieva et al. (2014) for the three atmospheric conditions

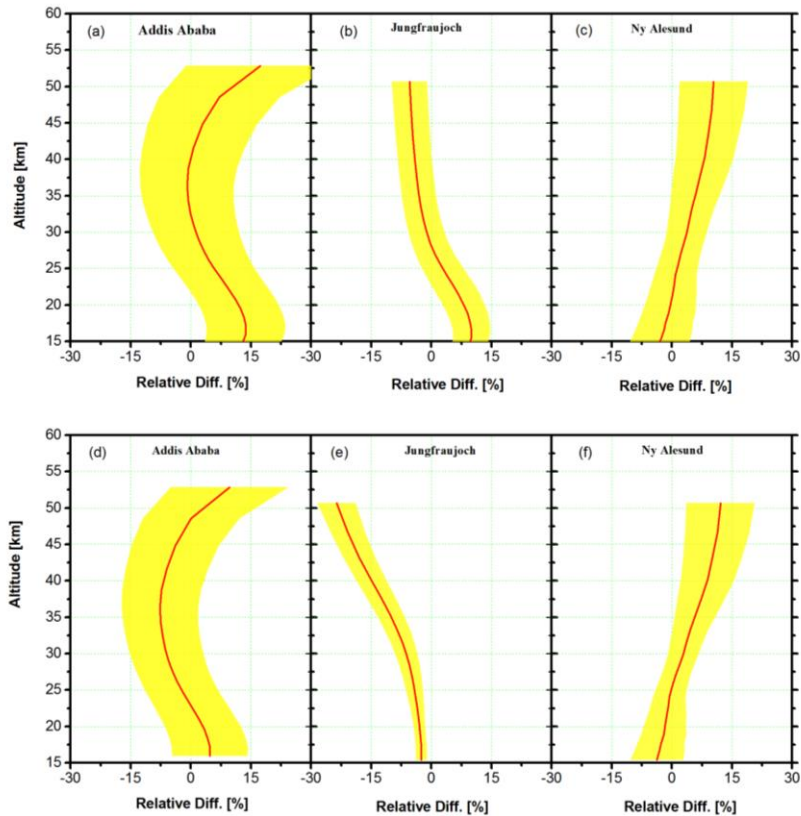
using at least two different measurement techniques. Furthermore, correlations analysis between $\text{CH}_4\text{-N}_2\text{O}$ measured by MIPAS and MIPAS CH_4 and MLS CH_4 has been used to show the variation of the uncertainty of MIPAS CH_4 as a function of latitude and altitude in a global scale. Finally, the cause of high uncertainty of MIPAS_ CH_4 _220 and its reduction in MIPAS_ CH_4 _224 at the lower stratosphere of tropics has been assessed through taking its relation with water vapour variability using a regression analysis method.

Inserted after the period in P5L14; “Both the estimated standard deviation (SD) of instrument uncertainty (i.e. MIPAS CH_4) and standard deviation of water variability for a given location, time of year, and layer were obtained using equations 4. Applying equation (4) to these data sources creates two sets of SD of MIPAS CH_4 uncertainty estimates. Similarly, SD of water vapour variability was obtained for each of the three latitudinal bands. The value estimated SD uncertainty of MIPAS CH_4 was calculated as square root of the mean variance estimates from the two data sources.”

The following paragraph has been added as a last paragraph under methodology section so that to make clear the methods employed in the manuscript.

Replace the last paragraph in P5L15-20 by “In addition to the above methods employed in this paper, as the UT/LS, mixing ratios of these long-lived trace gases are largely controlled by dynamical processes, generally resulting in compact tracer-tracer correlations. These correlations are usually more compact in high and mid-latitudes, while in tropics a somewhat larger scatter is observed (Plumb et al., 2007; Payan et al., 2009). We used such methods to show the variation of MIPAS_ CH_4 _220 uncertainty with high value at LS of tropics and its reduction in MIPAS_ CH_4 _224 as a function of latitude and altitude in a global scale using corresponding values MIPAS_ N_2O _220, MIPAS_ N_2O _224 for February 2010. In addition, both version data sets of MIPAS CH_4 and MLS CH_4 version 3.3 for February 2010 have been discussed too. These correlations are calculated on latitude bins space by 30° and on an altitude grid with 7 levels and spacing of 2 km.”

- Figures need to better describe what is shown and ALL plots and captions need revisions. e.g., Figure 1 shows (X - Y)/Z differences but doesn't say what X, Y, and Z are. Figures 4 needs a better colour scale, panel labels, y-axis label, larger fonts, etc. Figure 8 should plot CH_4 vs. H_2O , not H_2O vs. CH_4 . Take a careful look at quality of all the figures.



Response: The caption has been replaced by “Figure 1. Comparisons of MIPAS CH4 220 profile with FTIR (upper panel) and MIPAS CH4 224 profile with FTIR (lower panel). The relative differences $(200 * (FTIR \text{ VMR} - MIPAS \text{ VMR}) / (FTIR \text{ VMR} + MIPAS \text{ VMR}))$ averaged over Addis Ababa, Jungfraujoch and NyÅlesund sites. Shaded area is the Standard deviation of the mean relative differences.”

Response: We have corrected all the points the referee had pointed out on figure 4 after re-organizing the figures such as Fig. 4 only about exploring the variation of the uncertainty of MIPAS_CH4_220 and MIPAS_CH4_224 as a function of latitude and altitude and showing the reduction of uncertainty on MIPAS_CH4_224. Fig. 5 shows the uncertainty after removing the square root of water vapour variability variances from the amount of vmr values of CH₄ and N₂O using eq. 5.

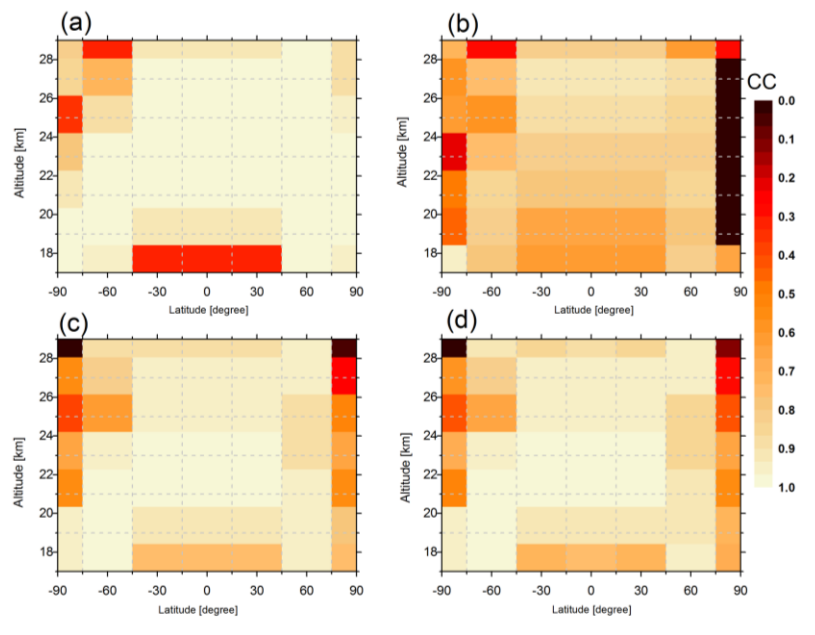


Figure 4: Correlation coefficients between (a) MIPAS_CH4_220 and MIPAS_N2O_220 (b) MIPAS_CH4_220 and MLS CH₄ (c) MIPAS CH₄ 224 and MIPAS_N2O_224 (d) MIPAS_CH4_224 and MLS CH₄ as a function of latitude and altitude for the period February 2010.

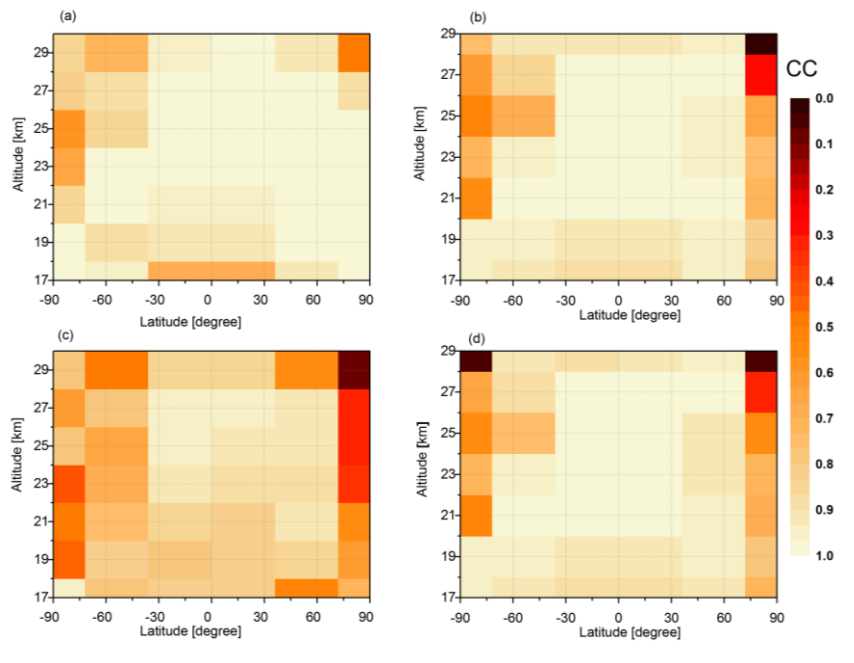


Figure 5: Correlation coefficients between (a) MIPAS_CH4_220 and MIPAS_N2O_220 (b) MIPAS_CH4_220 and MLS CH₄ (c) MIPAS CH₄ 224 and MIPAS_N2O_224 (d) MIPAS_CH4_224 and MLS CH₄ as a function of latitude and altitude for the period February 2010. After applying e.g.5 to remove the effect of water vapour variability on the latitudinal variation of the CC.

Figure 8. The random uncertainty of MIPAS CH₄ 220 (right) and MIPAS CH₄ 224 (left) versus the natural variability of H₂O using a three years data sets, 2009-2011 for altitude 18-21 km of tropics.

- *Is it correct to extrapolate the results from three specific sites as representative of the tropics, mid-latitudes, and polar regions? Justify this assumption.*

Response: The three sites are found at different atmospheric conditions, different regions. Hence, doing an atmospheric research at Addis Ababa mean that, we are discussing and presenting results that represent tropics. The latitudinal bands where we consider in this paper represent the three regions while the FTIR data were used. Here in this paper, we even taking a global scale a analysis of latitudinal variation of MIPAS CH₄ uncertainty and natural variability of water vapor (see Fig. 4 and Fig. 5).

- *Correlation seems be equated with cause, e.g., p16, para1.*

Response: The detection of high uncertainty of MIPAS_CH₄_220 in the lower stratosphere of tropics was related to water vapour variability as clearly seen in the correlation analysis which is Fig. 4 and Fig 5. Therefore, we have concluded that water vapour variability was the causal on the high uncertainty of MIPAS_CH₄_220.

- *Add a table stating sites/latitude bands used and time periods.*

Response: The tables in page 8 and page 9, which are results summery of the bias evaluation and scatter plot presented in this work. As you suggested, we improved the tables by adding a column that explains about latitudinal bands of the location, time period and number of coincidence are stated.

- *Use consistent terminology when referring to the MIPAS versions, e.g., both V5R_CH₄_220 and MIPAS CH₄ 220 are used. Use the former throughout, and similarly for V5R_CH₄_224.*

Response: corrected

- *Section 4.3 should be rewritten for clarity.*

Response: The description was indeed lacking detail interpretation of the method, some needed explanations in the main text. We added a new paragraph that can make it clear to understand the important of the section.

Tracer-tracer correlation (i.e. CH₄-N₂O) measured by MIPAS has been used to show the variation of the uncertainty of MIPAS CH₄ as a function of latitude and altitude. As revealed in different literatures (eg. Plumb et al., 2007; Payan et al., 2009), the correlation coefficients of long lived trace gases were also used to show the latitudinal and altitudinal variation of uncertainty of instrument. Figure 4. Show the correlation coefficients between MIPAS_CH₄_220 and MIPAS_N₂O_220, MIPAS_CH₄_220 and MLS CH₄, MIPAS_CH₄_224 and MIPAS_N₂O_224, and MIPAS_CH₄_224 and MLS CH₄. The correlation coefficients between MIPAS_CH₄_220 and MIPAS_N₂O_220 are only used to

show the latitudinal variations of MIPAS CH₄ 220 uncertainty. The larger the correlation of coefficient is the lesser the uncertainty and vice versa. We need only to show the uncertainty is different at different latitudinal bands. The correlation coefficient between MIPAS_CH4_220 and MIPAS_N2O_220 as a function of latitude and altitude are 0.30, 0.98 and 0.96 in the lower stratosphere over tropics, mid and high latitudes respectively. Nevertheless, the correlation coefficient between MIPAS_CH4_224 and MIPAS_N2O_224 are 0.62, 0.80 and 0.66. Hence, the result indicates the reduction of uncertainty of MIPAS_CH4_224 as its correlation in the lower stratosphere exceeds that of MIPAS_CH4_220.

The explanation written below has been placed in “4.3. Correlation plots of CH₄ and N₂O”

The influence of natural variability of water vapour on the uncertainty of MIPAS_CH4_220 and MIPAS_CH4_224 in the lower stratosphere of tropics with reduced effect on new version data. The contribution of water vapour variability to the large uncertainty of MIPAS_CH4_220 at lower stratosphere can be shown by the following assumption.

Assume that water vapour variability at the lower stratosphere of tropics has an effect on the amount of MIPAS CH₄ profile. The large uncertainty of methane derived from MIPAS instruments in lower stratosphere of tropics is due to water vapour variability and this has shown in Fig. 5 by taking in to account the amount of water vapour variability that enhance the profile of methane in tropics using the equation below. Hence, the true concentration amount of MIPAS CH₄ in the lower stratosphere is expressed as follows:

$$X_t = X_m - SD_{NV} \quad 5$$

Where X_t is the concentration amount after removing the effects of water vapour variability on the vmr amount of CH₄ and N₂O, X_m is the amount of methane obtained from the measurement and SD_{NV} is the square root of estimated natural variability of H₂O variance at upper troposphere and lower stratosphere (see Fig. 5).

The latitudinal variation of uncertainty of MIPAS_CH4_220 has been related with the variability of water vapour. However, the latitudinal variation of uncertainties of the new version data is reduced as water is jointly retrieved with methane. Fig. 5: shows the reduction of latitudinal variations of uncertainty after eq. 5 has been applied on the data sets. The correlation coefficient between MIPAS_CH4_220 and MIPAS_N2O_220 as a function of latitude and altitude after application of eq. 5, high variation of correlation coefficient has been reduced as shown in Fig. 5. Thus indicates the effect of water vapour variability on the uncertainty of MIPAS_CH4_220 in lower stratosphere of tropics.

- Data providers who are co-authors don't need to be thanked in the Acknowledgements.

Response: The acknowledgement has changed as follows:

We greatly acknowledge the MLS science teams for the satellite data used in this study. Special thanks go to Dr. samuel takele for his contribution on calibrating the spectra

measured by the FTIR at Addis Ababa. Finally, authors would like to thank Mekelle and Addis Ababa universities for the sponsorship and financial supports.

- *The References should be revised to ensure that they are correct and have consistent formatting. Some have incomplete information and several are old AMTD references (e.g., Laeng et al., 2015; Sepulveda et al., 2012). AMTD references are to manuscripts under review. These should be updated to the published AMT references.*

Response: The references have been corrected as follows:

Errera, Q., Ceccherini, S., Christophe, Y., Chabrillat, S., Hegglin, M. I., Lambert, A., Ménard, R., Raspollini, P., Skachko, S., van Weele, M., 15 and Walker, K.A.: Harmonization and Diagnostics of MIPAS ESA CH₄ and N₂O Profiles Using Data Assimilation; Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-245, 2016.

Errera, Q., Ceccherini, S., Christophe, Y., Chabrillat, S., Hegglin, M. I., Lambert, A., Ménard, R., Raspollini, P., Skachko, S., van Weele, M., and Walker, K.A.: Harmonization and Diagnostics of MIPAS ESA CH₄ and N₂O Profiles Using Data Assimilation; Atmos. Meas. Tech., 9, 5895–5909, doi:10.5194/amt-9-5895-2016, 2016

Laeng, A., Plieninger, J., von Clarmann, T., Stiller, G., Eckert, E., Glatthor, N., Grabowski, Haenel, N., Kiefer, M., Kellmann, S., Linden, A., Lossow, S., Deaver, L., Engel, A., Harvig, M., Levin, I., McHugh, M., Noel, G., and Walker, K.: Validation of MIPAS IMK/IAA methane profiles, Atmos. Meas. Tech. Discuss., 8, 5565–5590, doi:10.5194/amtd-8-5565 2015.

Laeng, A., Plieninger, J., von Clarmann, T., Stiller, G., Eckert, E., Glatthor, N., Grabowski, Haenel, N., Kiefer, M., Kellmann, S., Linden, A., Lossow, S., Deaver, L., Engel, A., Harvig, M., Levin, I., McHugh, M., Noel, G., and Walker, K.: Validation of MIPAS IMK/IAA methane profiles, Atmos. Meas. Tech., 8, 5251–5261, doi:10.5194/amt-8-5251-2015, 2015.

Sepúlveda, E., Schneider, M., Hase, F., Garcíã, O., E., Gomez-Pelaez, A., Dohe, S., Blumenstock, T., and Guerra, J., C.: Long-term validation 10 of total and tropospheric column-averaged CH₄ mole fractions obtained by mid-infrared ground-based FTIR spectrometry, Atmos. Meas. Tech. Discuss., 5, 1381–1430, 2012.

Sepúlveda, E., Schneider, M., Hase, F., Garcíã, O., E., Gomez-Pelaez, A., Dohe, S., Blumenstock, T., and Guerra, J., C.: Long-term validation of total and tropospheric column-averaged CH₄ mole fractions obtained by mid-infrared ground-based FTIR spectrometry, Atmos. Meas. Tech., 5, 1425–1441, doi:10.5194/amt-5-1425-2012, 2012

- *The manuscript needs line-by-line copy editing to correct the many typographical, grammatical, and technical errors.*

Response: Done

- *Overall, the manuscript is poorly written. It needs a complete rewrite for scientific clarity. I encourage all of the authors to review the next version carefully prior to resubmission*

Response: As you suggested, we have added and some re-arrangements of the manuscript by adding an extended sentences and paragraphs on the manuscript so that it attains scientific clarity. We have put it below the changes made on the manuscript and those that is not stated under the comments of the referee.