

Responses to referee 2: (received: 12 July 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 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.

Paper review

*This paper shows that water vapor variability impacted the methane concentration retrievals from MIPAS 220 and is improved in the 224 version. The paper uses a methodology of using coincident measurements from two data sources to derive the atmospheric variability and each instrument's random noise contribution following a technique published by Fioletov 2006. First here are some major issues I have with the paper. Given the basic assumptions the Fioletov method I believe is OK however I think the authors need to consider some possible limitations. One being that the instrument noise may depend on the concentration amount of methane due to forward model nonlinearities. This can be checked by correlating the retrieved amount against its reported uncertainty supplied by the MIPAS team. Secondly when deriving the instrument noise estimate from the coincident data, it would be interesting to compare that to the value supplied with the data set as a validation of the method. Neither of those were not done.*

Response: This paper has addressed the causes of large uncertainty of MIPAS\_CH4\_220 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\_224 had been reduced as water profile is jointly retrieved.

We added a sentence: The noise that is a random error of the instrument didn't have any relation to the amount of methane concentration on the upper troposphere and lower stratosphere.

*The presentation is not clear in many places and needs to be reworked a lot in order to publish this. Figure 4 for example shows a before and after like correlation analysis for methane version 220. In the after figure the authors say they subtracted water vapor variability from the CH4 retrieval and show how much better the correlation has improved. I have no idea how you can after the fact remove the water vapor variability impacts from the 220 data set or the 224 (figure 5) data set (which itself is significantly improved in this regard due to the simultaneous retrieval of H2O and CH4). Because the authors provide no explanation of how this is done, I am recommending rejection. The "removing" the effect of H2O interference leading to greatly improved agreement with correlative data is the central*

*point of the paper and establishment of cause described in the title, this needs to be much better explained*

Response on Page 9 lines 16–20: The contribution of water variability on the uncertainty of MIPAS CH<sub>4</sub> has clearly seen in figure 4, we showed that the correlation between the two tracer gases derived from the instruments are expecting not to vary with latitude. However, the figure shows the correlation coefficient is less at lower stratosphere of tropics. On the other hand, this result confirms the large uncertainty of MIPAS\_CH4\_220 at lower stratosphere of tropics as obtained from bias evaluation and is due to the effects of atmospheric water variability.

The explanation written below has been placed in “4.3. Correlation plots of CH<sub>4</sub> and N<sub>2</sub>O”

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 CH<sub>4</sub> 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 CH<sub>4</sub> 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 CH<sub>4</sub> and N<sub>2</sub>O,  $X_m$  is the amount of methane obtained from the measurement and  $SD_{NV}$  is the square root of estimated natural variability of H<sub>2</sub>O 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.

*The text was hard to follow and comprehend and I give a few examples of this below.*

*On page 9 line 18 there is a reference to a middle panel in figure 4 a figure with 4 panels in a 2X2 arrangement. What is the middle panel?*

Response: The error has been created while we taking the first figure prepared in the draft with three panel.. Now, it has been corrected.

*Figure 7 show profiles of H<sub>2</sub>O variability at three station sites. The profile at some altitudes is clipped at zero suggesting that it either is negative (not possible) or unknown. One profile it is exactly zero which is extremely unlikely (ie absolutely no atmospheric variability).*

Response on Page 12 lines 6-8 and page 13 figure 7: Fig. 7 shows the natural variability of H<sub>2</sub>O determined using Eq. 4 from the data MIPAS V5R\_H<sub>2</sub>O\_220 and MLS (Ver3.3) for the time period of February 2010. The resulting natural variability at 15-17 km altitude are 8.4 %, 5.4 % and 3.4 % for Addis Ababa, Jungfraujoch, and NyAlesund. Here in the figure the variability of water vapour becomes negative that shows an over estimation of random errors of the instruments are exist as the natural variability is a combination of the three estimated sample variances and had large uncertainty (see Eq. 4). Moreover, natural variability of water vapour could also find from the MIPAS instrument using

*I don't know why the figure 8 scatter plot of monthly averages in one height range for the 224 data set contains many more the 36 points for 3 year monthly averages*

Response: In our previous manuscript, we used averaged on each level such as 18, 19, 20 and 21 km. now corrected as averaged values between 18-21km

The following sentence has been added in page 14 L10: The number of points is 36 in both scatter plot that represent the monthly averaged SD natural variability of water vapour and uncertainty of MIPAS\_CH<sub>4</sub>\_220 and MIPAS\_CH<sub>4</sub>\_224 in the altitude ranges of 18-21 km. for time period of three year.

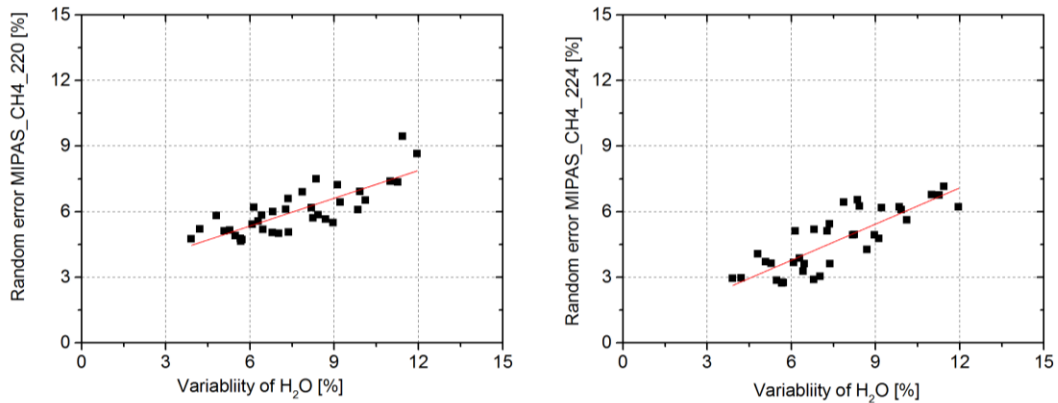


Figure 8: The scatter plot between standard deviation of natural variability of H<sub>2</sub>O and the random uncertainty of MIPAS\_CH4\_220 (right) and MIPAS\_CH4\_224 (left) using a three years data sets, 2009-2011 for altitude 18-21 km and latitudinal band of tropics in the lower stratosphere.

*A sentence on page 13, line 15 seems to refer to a figure not included in the paper. It cannot possibly be describing figure 8.*

Page 13 lines 15: While we prepared this manuscript, we were trying to put the relation between the uncertainty of MIPAS CH<sub>4</sub> and variability of water vapour using a correlation coefficient between SD uncertainty of MIPAS CH<sub>4</sub> and SD of water vapour variability as function of altitude. Finally, we decided to put it in scatter plot at the lower stratosphere of tropics.

*There are a lot wording and grammer errors in here that need improving.*

Response: we have taking in to consideration all the referee has suggested for improving the manuscript, such major corrections has been posted below.

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 CH<sub>4</sub> 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 CH<sub>4</sub> 224 methane were obtained 2.4 %, 1.4 % and 5.1 %. Moreover, the correlation coefficient between MIPAS CH<sub>4</sub> 220 and MIPAS V5R\_N<sub>2</sub>O\_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 CH<sub>4</sub> 224 and MIPAS V5R\_N<sub>2</sub>O\_224 are 0.62, 0.80 and 0.66.

P3L2-5: the sentences have been replaced by “The coincident measurements Of H<sub>2</sub>O, CH<sub>4</sub> and N<sub>2</sub>O 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 H<sub>2</sub>O. MLS CH<sub>4</sub> was derived from EOS MLS coincident measurements of atmospheric water vapour (H<sub>2</sub>O), carbon monoxide (CO) and nitrous oxide (N<sub>2</sub>O).”

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 (CH<sub>4</sub>) 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 CH<sub>4</sub>-N<sub>2</sub>O measured by MIPAS and MIPAS CH<sub>4</sub> and MLS CH<sub>4</sub> has been used to show the variation of the uncertainty of MIPAS CH<sub>4</sub> as a function of latitude and altitude in a global scale. Finally, the cause of high uncertainty of MIPAS\_CH4\_220 and its reduction in MIPAS\_CH4\_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<sub>4</sub> ) 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<sub>4</sub> 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<sub>4</sub> 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\_CH4\_220 uncertainty with high value at LS of tropics and its reduction in MIPAS\_CH4\_224 as a function of latitude and altitude in a global scale using corresponding values MIPAS\_N2O\_220, MIPAS\_N2O\_224 for February 2010. In addition, both version data sets of MIPAS CH<sub>4</sub> and MLS CH<sub>4</sub> version 3.3 for February 2010 have been discussed too. These correlations are calculated on latitude bins space by 30<sup>0</sup> and on an altitude grid with 7 levels and spacing of 2 km.”

