

Responses to referee 1: (received: 27 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 and reorganized the manuscript, in many parts extended sentences and 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 of 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. The page number and lines indicated on the lists are taken from commenting manuscript.

This paper presents retrievals of CH₄ and N₂O using a ground-based FTIR instrument at Addis Ababa, Ethiopia with aim to present observations, error analysis, and comparisons with satellite data. The lack of long-term remote sensing observations at Addis Ababa makes this work important. The technique and results of such measurements might be interesting and likely suitable for the journal. However, I have major comments and foremost revisions are warranted before publication. In my opinion, the quality of the paper needs to be improved before publication.

Response: We would like to thank the reviewer for this positive evaluation and critical comments that would help us to make the paper more vital to the scientific community. Furthermore, the response of each major comments and specific comments will place following each comment.

Here, some of the figures we have shown in this paper are important as the article is the first result of FTIR CH₄ and N₂O, such figures would be ignored while we prepared other related works. Some responses explained below are related to the work done here is the first for this site.

Major Comments

I have the following major comments:

- (1) *It is not clear to me what exactly is (are) the goal(s) the authors try to achieve. There is a lack of description in both the FTIR measurements and comparison with satellites. The authors need to specify and emphasize what is (are) the goal(s). Is the goal to present a retrieval strategy of CH₄ and N₂O? or compare/validate three satellites with the ground-based FTIR?. In the manuscript, it is mentioned that a satellite is used to validate the FTIR, which is quite surprising. Normally, high-resolution FTIRs are used to validate satellite retrievals. In general, the manuscript is short and lack important details in many sections, e.g., FTIR measurements, satellite, and results.*

Response: The goals of this paper are retrieval strategy of CH₄ and N₂O, since the micro windows applied here are somewhat modified. Furthermore, validations of the ground based FTIR CH₄ and N₂O have been made. As the referee has stated, FTIR is a high resolution and used to validate the satellite observations. However, the FTIR at Addis Ababa is new and it required to verify the measured parameters through validation.

(2) The retrieval strategy applied in this work is different than the NDACC/IRWG recommendations. I highly suggest to try the harmonized NDACC suggested retrievals and compare with your results. In particular, micro-windows applied here are different that NDACC recommended micro-windows. Furthermore, Sussman et al. (2011) found that HDO is important interfering specie. However, here is not included or even mentioned.

Response: The reasons why we modify the micro windows are due to high residuals obtained between the measured and synthesized spectra at the Addis Ababa site. Thus, the micro-windows recommended by the NDACC might be useful for the other FTIR sites found at mid and high latitudes. The Micro windows applied here are different from the micro windows used by Sussman et al. (2011) and that is why the HDO is not an important interfering species.

(3) The DOFs obtained from FTIR measurements is limited to a value of approximately 2. Hence, the typical information content will allow to retrieve tropospheric and stratospheric columns. However, the authors show a comparison of profiles with satellite, which are mainly sensitive in the stratosphere. Furthermore, comparisons are carried out using a limited number of years, even though measurements at Addis Ababa started in 2009. Additionally, the criteria to establish coincident measurements between FTIR and satellite needs to be revised.

Response: As the referee stated, the DOFs obtained from FTIR measurements are 2. Due to the limitation of the sensitivities of satellite that is upper troposphere and stratosphere, the comparisons have been done on the stratosphere to increase the number of coincident days. During the measurements of FTIR, there are several days and months where the instruments were not functional. To make it more clear the period time that does not have a measured value will be stated (added) as a new description in section 2.1.

Specific Comments

I highly suggest to review exhaustively the English along with the manuscript. I have some specific comments below, but they are not exhaustive by any means. P1, L2: Change Addis Ababa with Addis Ababa, Ethiopia

Response: Done

Is the instrument/site part of the NDACC effort?, if not please explain the reason.

Response: The instrument or site is not part of NDACC yet. Since all the measured species have not yet validated, only ozone and water vapour are validated by the previous PhD student (Dr. samual T.). This article was prepared to register the site as members of NDACC and other works has also submitted.

Are the measurements automated?, how often do you measure.

Response: No, we measured them starting from May 2009 to February 2013 with some days and months are missing (See Section 2.1).

Add information about quality control of spectra acquired.

Response: We have added the following sentence about the quality of the acquired spectra at the end of Section 2.2.

The quality of the measurements during the time period of May 2009-Feberoury 2011 has revealed by Takele Kenea et al., (2013).

Change ground based with ground-based when mentioned in the manuscript

Response: Done

P3 l13. Change “The Addis Ababa FTIR spectroscopy” for “The ground-based FTIR at The Addis”

Response on P3 L13: Done

P3, L15 add Altitude of the site

Response on P3 L15: We added “, 2443 m a.m.l.” after 38.76°E.

P.3, Section: In the measurement site section is not clear whether the site is located in the city limits of Addis Ababa or whether is located far from major emission sources. Please add information regarding typical air masses transported at Addis Ababa and/or emitted from local sources. Also, are there other atmospheric measurements carried out in Addis Ababa, which can be used to complement your study?.

Response on P3, Section 2.1: As suggested by the referee, we have added sentences that describe the details of the measurement site, Addis Ababa and the period time when the

instrument was operational. As far as I know, no other atmospheric measurements are carried on in Addis Ababa.

P3, L23: remove very

Response on P3, L23: Done

P3, L23, change “to the study of trace gases in the atmosphere” with “to study trace gases in the atmosphere”

Response on P3, L23: Done

P3, L23, remove “terrestrial”

Response on P3, L23: Done

P3, L30: remove very

Response on P3, L30: Done

P4, L7: explain why Tikhonov-Phillips regularization was used and also why is the retrieval performed on a logarithmic scale.

Response on P4, L7: Since the distribution of the a priori profile of the species has a logarithmic or exponential distribution in the upper troposphere and lower stratosphere over Addis Ababa.

P4, L29: The link <http://www2.cesm.ucar.edu/working-groups> does not work. Similarly, the link: <http://hyperion.gsfc.nasa.gov/Dataservices/automailer/index.html>. Consider changing and being consistent with format.

Response on P4, L29:

P5, L1: Why different versions of HITRAN were used. Please explain and also, what versions were used for gases

Response on P5, L1: As the referee suggested, we added sentences that description which versions of HITRAN data were used for gases. Different versions of HITRAN were used as the new updated HITRAN.....

Required

P5: I suggest to remove Figure 1. I do not see the value of Figure 1. It does not show a result/finding but only the a priori profiles used. Furthermore, in the text the temperature profile from this figure is not even mentioned. Keeping the text would be ok just change it accordingly.

Response on P5: We agree and removed it. Similarly, the text has been changed as follows.

Both methane (CH₄) and nitrous oxide (N₂O) are well-mixed in the troposphere and their VMR decrease with height and becomes negligible with no variation above 55 km. The vertical variability of N₂O and CH₄ in the lower stratosphere is characterized by a large vertical gradient

P5. What do you mean by “The micro windows have been adopted from different sources.”, do you mean past works? Expand a description and add references.

Response on P5: We mean, micro windows that give us a minimum residuals and errors were considered. The references from where the micro windows are adopted has been added at P5, L8.

The microwindows have been adopted from different sources (.Senten et al. 2008; Sussmann et al., 2011; Arndt et al., 2004).

P5. L8-10. It is mentioned that micro windows are different than NDACC /IRWG guide-lines. Please describe in detail why the NDACC guideline was not adopted. I was able to retrieve the guideline and all micro-windows for CH₄ and N₂O used in this work are different. I imagine the guidelines were created to obtain a harmonized retrieval strategy applied among different locations. This is important and needs to be explained.

Response on P5, L8-10: The reasons why we modify the micro windows are due to high residuals obtained between the measured and synthesized spectra at the Addis Ababa site. Thus, the micro-windows recommended by the NDACC might be useful for the other FTIR sites found in mid and high latitudes.

In order to characterize the possible impact of the choice of the micro windows I suggest to test and compare the micro-windows/setting applied here with the harmonized NDACC settings.

Response: We have done using the harmonized NDACC settings and comparing them with those used in this paper. However, the comparisons are not stated in the paper.

Table 1. Recommendation: Change T.Gases with Gas; replace parenthesis with dash for micro-windows, change int. gases with interfering species; why is important to show three significant figures in DOF?, and mention here what species are retrieved as profiles and columns.

Response on Table 1: As suggested by the referee, we agree that the title of the Table has to be clear; T. Gases have been changed to “Gas”, MW to “micro-windows”, int. gases to “interfering species”.

Table 1. According with the NDACC/IRWG guideline, HDO is an interfering specie but you are not using it, please explain. See also Sussmann et al. (2011).

Response: As we have stated previously, the micro-windows used in this paper are different from the micro-windows recommended in the NDACC / IRWG guideline that have been applied to other atmospheric conditions.

Along the same lines, water vapor might influence the retrieval of CH₄ and N₂O. However, it is not mentioned what water vapor profile is used. Please describe if climatology (or reanalysis) is used, or do you pre-retrieve water vapor?.

Response on P5, Table 1: No, We use WACCM reanalysis data

P5, L9: *change wondows with windows*

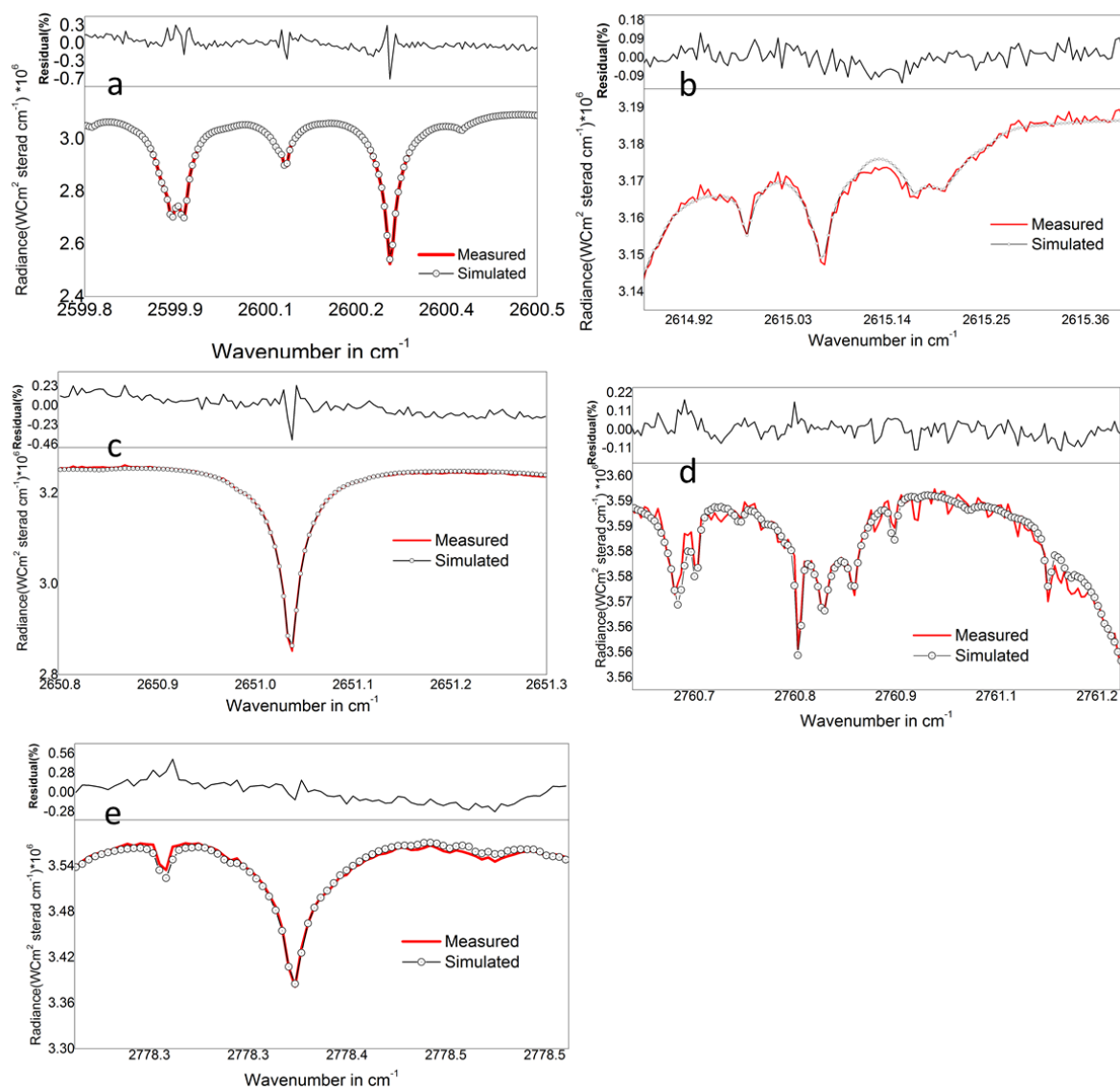
Response on P5, L9: Done

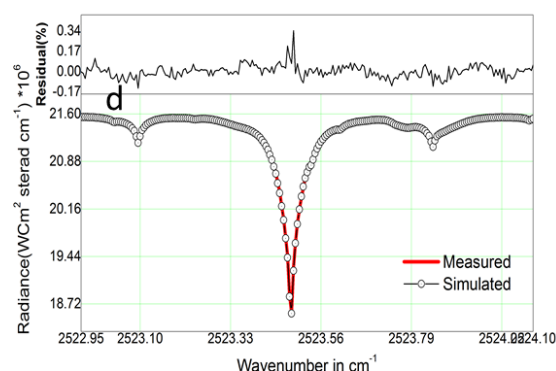
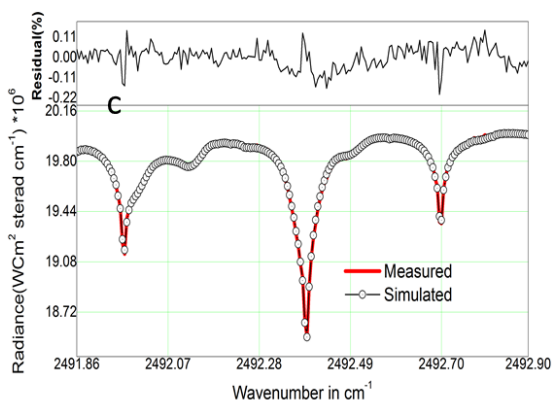
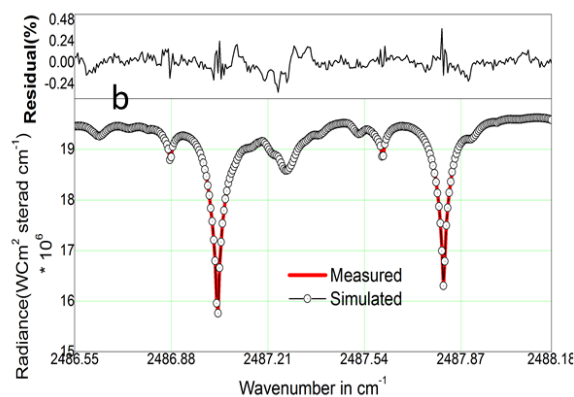
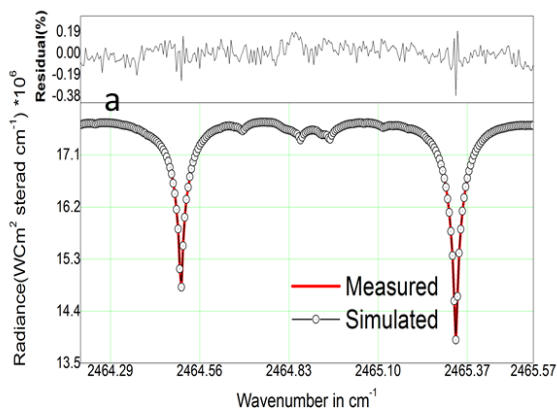
P5, L10: *The link www.ndacc.org does not have information regarding retrievals of CH₄ and N₂O. Change it accordingly.*

Response on P5, L10: We have changed it adding the following instead of www.ndacc.org “the EU projects UTFIR (www.nilu.no/uftir) and HYMN (www.knmi.nl/samenw/hymn) reports.”

Quality of Figures 2 and 3 is not good enough. They are blurry and too small to really see the quality of the fits.

Response on Figures 2 and 3: Those two figures have been updated as follows:





P5, L11. I suggest to split the following sentence: The spectral fit and residual between measured and simulated spectra at five and four microwindows for CH₄ and N₂O respectively are depicted in Fig. 2 and Fig. 3 for example spectra recorded on Feb 26, 2013 and Dec 31, 2009 at Addis Ababa respectively.” And why two different dates are used?, and remove Addid Adaba, you are not using other sites.

Response on P5, L11: Since we prepared the retrieval and validation results of both CH₄ and N₂O separately. The sample spectra presented in the paper for CH₄ and N₂O were different. The sentence has also separated into two sentences.

The spectral fit and residual between measured and simulated spectra at five micro windows for CH₄ is shown in Fig. 2 for spectra recorded on Feb. 26, 2013. Whereas, N₂O uses four micro windows is depicted in Fig. 3 for spectra recorded on Dec 31, 2009.

P5, L15: What do 0.4% and 0.35% mean?, explain.

Response on P5, L15: To make clear why we have put those results in the paper, the following paragraph would be added.

Generally residual of the spectra mean that the difference between measured and synthesis spectra. Furthermore, the residual can also be expressed in percentage while we took the ratio of the difference and measured times 100. This residual was used to explain the quality of the measured spectra which we have used to derive the concentration or amount of both CH₄ and N₂O.

P5, L15. It is mentioned that the retrieval strategy is optimized using a single spectra: : : please expand this description, what do you use a criteria for optimization? Is it consistent for all months, zenith angles?.

Response on P5, L15: The optimization of the retrieval strategy starts from the selection of the micro windows which are somewhat different from the recommended by NDACC.

Regarding figure 4. Averaging kernel matrices are not described in the text. I recommend to remove the matrices and keep the rows of the averaging kernels. Rows of averaging kernels are not visible within the x-axis scale, please re-adjust. The sum of averaging kernels can be divided by 10 to use the same x-axis scale and lines need to be color coded by altitude and show the color bar. As all other figure, increase the quality of the figure.

Response Regarding figure 4:

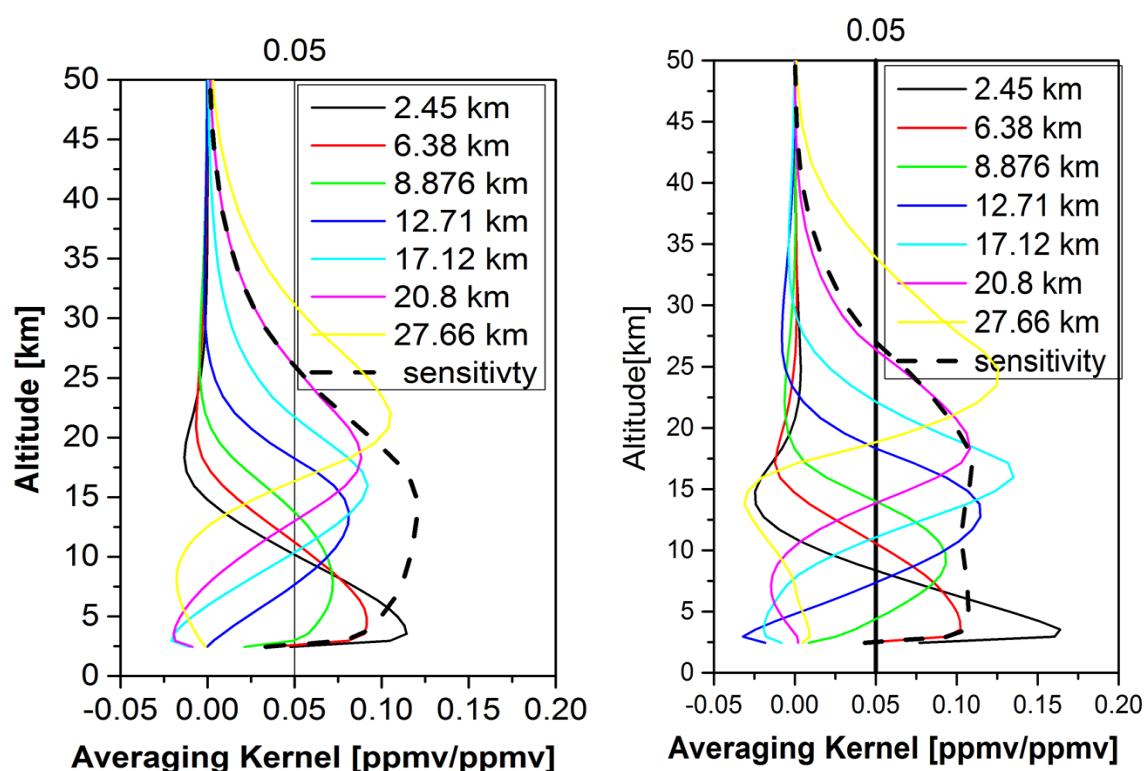


Figure 3. Sensitivity analysis of the retrieved profiles of CH₄ (left) and N₂O (right) at Addis Ababa using the selected rows of the averaging kernels as a function of altitude. The dotted lines are the sum of the rows of the averaging kernels for a spectra measured on Feb. 26, 2013 for CH₄ and Dec 31, 2009 for N₂O.

P7, L12. Along the manuscript Addis Ababa is mentioned, although is clear. I suggest to remove Addis Ababa when is not needed.

Response on P7, L12: The Addis Ababa has been removed from the following specific locations in the text.

P9, L10. *Examples of error profiles are shown for CH₄ - Feb 26 2013 and N₂O – Dec 31 2009. Why was decided to use examples that are +3 years apart?*

Response on P9, L10: Because we first prepared all the analysis separately for CH₄ and N₂O

Figure 5. Please improve the quality of the figure. It is extremely hard to find the corresponding error type. I suggest to add the total error for each statistical and systematic errors. Additionally, I don't follow why the retrieved/apriori profiles are shown here. I suggest to replace this by the fraction of the total error with the retrieved profile and one might see the fractional error as a function of altitude.

Response on Figure 5.: We have improved the quality of the figure according to the suggestion of the referee and we added the following expressions about the fraction total error.

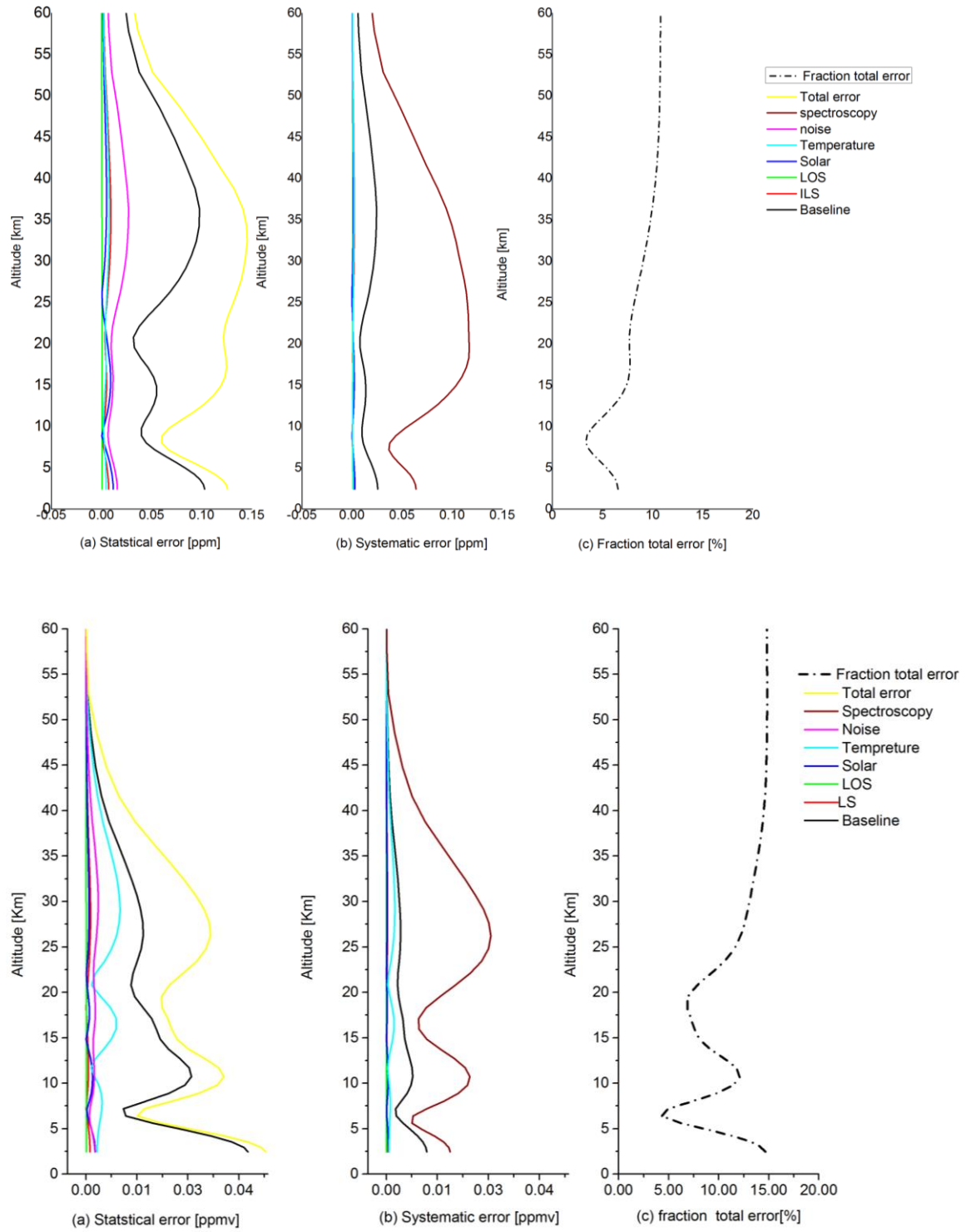


Figure 4 Estimated errors for the profiling retrieval of CH₄ (Top) and N₂O (bottom) over Addis Ababa: (a) statistical (random) errors (b) systematic errors of parameter listed in the legends, (c) Fractional total error [%].

P10, L7. It I mentioned that the MIPAS reduced spectral resolution is used, what does reduced resolution mean?

Response on P10, L7: The measurements collected between January 2005 and April 2012 it measured with a reduced spectral resolution (RR, theoretical resolution: 0.0625 cm^{-1} , apodised resolution: 0.121 cm^{-1}) but with a finer tangent altitude spacing. The data used in this paper is

P10, L8. Explained why only satellite data between March 2009 to Dec 2010 is used for MIPAS?, what about the other satellite measurements (it is not mentioned)? why this wide range is used. I would try other distances as well. How do you assess the spatial-temporal variability of both CH_4 and N_2O .

Response on P10, L8: We have added the statement to explain the period time used to validate FTIR with MLS.

The comparison of FTIR with MLS for a period time of May 2009 to February 2013 has also made. Here, in this paper, we did not put anything about the spatial-temporal variability of both CH_4 and N_2O .

P11, L2. What does visibility flag 1 mean?, and how the criteria of diagonal elements has been chosen?

Response on P11, L2: The visibility flag indicates whether spectral data was available for the given altitude (value=1) or not (value=0). Altitudes with visibility flag = 0 have to be omitted. In this paper, we have taken data with visibility flag value of 1. Similarly, the diagonal element of the averaging kernels that indicates the sensitivity of the instrument and its value above 0.03 has been taking on this work.

P11, L5. Change Plieninger et al. 2016 with Plieninger et al. (2016) and check format of references along the manuscript.

Response on P11, L5: Done

P11, L17. Under the MLS section it is mentioned: “In this work, we have used version 3.3 MLS of N_2O data set to validate ground-based FTIR results”. This is kind of surprising. Usually, the ground-based FTIRs are used to validate satellite-based measurements. Please explain in detail why you have chosen MLS to validate FTIR. Also, do you also use MIPAS and AIRS to validate FTIR?

Response on P11, L17: The results obtained from the ground-based FTIR observations at Addis Ababa are presented and discussed in this paper for the first time to this latitude band. This is the reason why we verify the quality of the FTIR measurements by MIPAS, MLS and AIRS.

P11, L19, Change EOS MLS (Earth Observing System) with EOS MLS

Response on P11, L19: Done

P12, L2. Expand the description “Selection criteria were implemented as stated in Livesey et al. (2013)”. What do you mean by selection criteria?

Response on P12, L2: The selection criteria mean that the

In order to see the difference in sensitivity among the satellite measurements I suggest to include averaging kernels for the three selected satellites.

Section 5.1. The coincident criteria of 2 deg latitude and 10 deg of longitude from the FTIR site is extremely large. As in other parts of the manuscript, please expand a description of

Section 5.1. I encourage the authors to rename the following: V5R_CH4_224, V5R_N2O_224, MLS V3.3. These names are constantly but highly distractive. P13, eq 4. It might be obvious but please describe variables of eq 4.

Response, Section 5.1.: The paragraph and equations have been rewritten as follows; $\text{sat}_i(z)$ has changed to $X_s(z)$ in equation 4.

The ground based FTIR measurements of CH₄ and N₂O has been validated at different locations (e.g. Senten et al. 2008). MIPAS, MLS and AIRS have a better vertical resolution than ground-based FTIR profiles and high temporal and spatial coverage in the tropics. The analysis of the comparison between volume mixing ratio values derived from FTIR and MIPAS were performed for the data sets between March 2009 to December 2010. Furthermore, The comparison of FTIR (CH₄, N₂O) with a MLS (CH₄, N₂O) and AIRS (CH₄) for the time period of May 2009 to February 2013 has also made. Hence, the profiles from MIPAS, MLS and AIRS have been degraded to make a comparison between the FTIR and satellite observations. Therefore, the satellite measurement profiles are smoothed using the FTIR is averaging kernels of individual species obtained from the ground based FTIR retrieval by applying the procedures reported in Rodgers and Connor (2003 and given as

$$X_s = X_a + A(X_i + X_a)$$

The absolute difference at each altitude layers of a pair profile is calculated using

$$\delta_i(z) = [FTIR_i(z) - X_s(z)]$$

Where $FTIR_i(z)$ and $X_s(z)$ are the FTIR and smoothed satellite profiles of CH₄ or N₂O respectively. The mean squares error can be expressed as

$$MSE_i = \sqrt{\frac{1}{N(z)-1} \sum_{i=1}^{N(z)} \delta_i(z)^2}$$

$$\Delta_{rel}(z) = \frac{1}{N(z)} \sum_{i=1}^{N(z)} \delta_i(z)$$

Where $\delta_i(z)$ is the difference (absolute or relative), $N(z)$ is the number of coincidences at z , $FTIR_i(z)$ is the FTIR VMR at z and the corresponding $X_s(z)$ smoothed volume mixing ratio derived from satellite instruments.

P13, eq 10. Why is this equation multiplied by 200?. Please revise this and all other equations.

Response on P13, eq.10: We multiply it by 200 because the denominator was multiplied by 0.5 and we corrected it as follows:

$$RDiff(z) = \frac{100 * [PC_{FTIR}(z) - PC_{sat}(z)]}{[PC_{FTIR}(z) + PC_{sat}(z)]/2}$$

Section 5.2. It is not explained why authors compare FTIR vs satellite vertical profiles. The FTIR information content is limited to 2 DOFs (tropospheric and stratospheric columns) but main figures for the comparison are shown as profiles.

Response, Section 5.2.: Since those results are the first for Addis Ababa.

References Sussmann, R., Forster, F., Rettinger, M., and Jones, N.: Strategy for high-accuracy-and-precision retrieval of atmospheric methane from the mid-infrared FTIR network, *Atmos. Meas. Tech.*, 4, 1943-1964, <https://doi.org/10.5194/amt-4-1943-2011>, 2011.

Response: corrected