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# Methane and nitrous oxide retrievals from MIPAS-ENVISAT

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#### Abstract

We present the strongly revised IMK/IAA MIPAS-ENVISAT  $CH_4$  and  $N_2O$  data products for the MIPAS full resolution (versions V5H\_CH4\_21 and V5H\_N2O\_21) and for the reduced resolution period (versions V5R\_CH4\_224, V5R\_CH4\_225, V5R\_N2O\_224 and

- <sup>5</sup> V5R\_N2O\_225). Differences to older retrieval versions which are known to have a high bias are discussed. The usage of the HITRAN 2008 spectroscopic dataset leads to lower values for both gases in the lower part of the profile. The improved correction of additive radiance offsets and handling of background radiance continua allows for aerosol contributions at altitudes in the upper stratosphere and above. These changes
- <sup>10</sup> lead to more plausible values both in the radiance offset and in the profiles of the continuum absorption coefficients. They also increase the fraction of converged retrievals. Some minor changes were applied to the constraint of the inverse problem, causing small differences in the retrieved profiles, mostly due to the relaxation of off-diagonal regularisation matrix elements for the calculation of jointly retrieved absorption coeffi-
- cient profiles. Spectral microwindows have been adjusted to avoid areas with saturated spectral signatures. Jointly retrieving profiles of water vapour and nitric acid serves to compensate spectroscopic inconsistencies. We discuss the averaging kernels of the products and their vertical resolution.

### 1 Introduction

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS, Fischer et al., 2008) is a Fourier transform spectrometer on board of the Environmental Satellite (ENVISAT). This satellite was launched in March 2002 and operated by the European Space Agency (ESA) until April 2012, when it ceased all communication to ground. During that time, MIPAS measured atmospheric emission spectra in the infrared between 685 and 2410 cm<sup>-1</sup> in limb geometry. There are two MIPAS measurement periods:



during the full resolution period from June 2002 until March 2004, the instrument mea-

sured with the originally specified spectral resolution of  $0.025 \text{ cm}^{-1}$  (0.0483 cm<sup>-1</sup> after an apodisation). Due to a malfunction in the interferometer slide system, there is a data gap until January 2005 when the instrument recommenced measuring with a so-called reduced resolution of  $0.0625 \text{ cm}^{-1}$  (0.121 cm<sup>-1</sup> after the apodisation). The instrument

- <sup>5</sup> has been operated in several different measurement modes with different tangent altitude patterns. For this study, only spectra from the MIPAS nominal measurement mode have been used; this covers about 80 % of the total measurement time. For the full resolution period, one limb scan contains 17 different spectra with tangent altitudes from 6 to 68 km. A limb scan in the reduced resolution period consists of 27 different spectra
- <sup>10</sup> and their tangent altitudes are latitude dependent. The lowest tangent altitude ranges from 6 km at the polar regions to 9 km at the equator and the highest tangent altitude increases from 70 km in polar regions to 73 km at the equator.

The processor developed at the Institute of Meteorology and Climate Research in cooperation with the Instituto de Astrofísica de Andalucía (CSIC) retrieves profiles of

temperature and various trace gases. The retrieval of  $CH_4$  and  $N_2O$  with this processor was first described in Glatthor et al. (2005) for the full resolution period and in von Clarmann et al. (2009) for the reduced resolution period.

Various studies showed  $CH_4$  and  $N_2O$  profiles retrieved from MIPAS-ENVISAT to have a positive bias in the lower part of the profiles. In Glatthor et al. (2005) the largest

- <sup>20</sup> CH<sub>4</sub> and N<sub>2</sub>O values are compared to tropospheric climatological data and found to be 0.7 ppmv higher for CH<sub>4</sub> and 80 ppbv higher for N<sub>2</sub>O. Höpfner et al. (2007) compared MIPAS full resolution CH<sub>4</sub> and N<sub>2</sub>O profiles versions V3O\_CH4\_9 and V3O\_N2O\_9 to those measured by ACE-FTS in version 2.2. This study uses profiles measured during northern polar winter. Below 15 to 18 km, MIPAS CH<sub>4</sub> profiles showed a high bias
- of around 0.2 ppmv while for N<sub>2</sub>O a MIPAS bias of around 25 ppbv was found below 15 to 18 km. von Clarmann et al. (2009) state that the bias in the reduced resolution period is smaller than in the full resolution, but still present. Laeng et al. (2015) did a detailed validation of MIPAS reduced resolution V5R\_CH4\_222, comparing to profiles from ACE-FTS, SCIAMACHY, HALOE, SOFIE, Mark IV balloon interferometer and



cryosampler in situ measurements. They found MIPAS profiles to be biased high below 25 km by about 0.2 ppmv.

The aim of this study has been the improvement of the MIPAS IMK/IAA  $\rm CH_4$  and  $\rm N_2O$  products.

#### **5 2 General retrieval descriptions**

discussed.

Here we discuss the latest retrieval setup of CH<sub>4</sub> and N<sub>2</sub>O. For the full spectral resolution period this refers to data versions V5H\_CH4\_21 and V5H\_N2O\_21 and for the reduced resolution period to data versions V5R\_CH4\_224, V5R\_CH4\_225, V5R\_N2O\_224 and V5R\_N2O\_225. For simplicity, in the following only the last part
of the version code will be used. This simplification, however, is not expandable beyond this work because version identifiers are gas specific. Versions 224 and 225 are almost identical. Version numbers are different only to guarantee complete traceability. The only technical difference is the source of the ECMWF analysis used to constrain the preceding temperature retrievals. This has no discernible effect on the CH<sub>4</sub> and N<sub>2</sub>O profiles. Thus, we do not discuss these versions separately. The data sets of these versions are disjoint in a sense that one observation is either 224 or 225. Thus the data sets are complementary. All this applies also to older pairs of versions i.e. 220 and 221, as well as to 222 and 223. Table 1 offers an overview of the data versions

<sup>20</sup> Volume mixing ratios (vmr) of  $CH_4$  and  $N_2O$  are retrieved in MIPAS channel B in several microwindows between 1220 and 1320 cm<sup>-1</sup> in the P-branch of the  $v_4$  band of  $CH_4$ with the MIPAS-ENVISAT data processor developed at IMK and IAA (von Clarmann et al., 2003). The level 1b data used for the retrievals was version MIPAS/5.02–5.06.

For each MIPAS limb scan profiles of  $CH_4$  and  $N_2O$  are retrieved. Since there are strong cross-interferences between the  $CH_4$  and the  $N_2O$  lines, both species are retrieved simultaneously in one retrieval step to minimise mutual error propagation. In addition, mixing ratios of the interfering species HNO<sub>3</sub> and H<sub>2</sub>O are jointly fitted to im-



prove the spectral residual. The profiles of temperature, pressure and ozone as well as the spectral shift correction are known from previous retrieval steps and are not treated as variables in the  $CH_4$  and  $N_2O$  retrieval, but their retrieved profiles are used within the radiative transfer calculations. An additive radiance offset correction is re-

- <sup>5</sup> trieved for each microwindow. It is constant for all tangent heights. Additionally, for each microwindow continuum absorption coefficient profiles are retrieved that account for continuum spectral contributions of atmospheric aerosol, uncertainties in the modelling of the continua of O<sub>3</sub>, H<sub>2</sub>O and N<sub>2</sub> and contributions of distant lines which sum up to a quasi-continuum (von Clarmann et al., 2003).
- A first order Tikhonov finite differences constraint (e.g. Tikhonov, 1963; Steck and von Clarmann, 2001) is used to fight ill-posedness and to reduce vertical oscillations. The a priori profiles for both gases are zero. Therefore, not the mixing ratios themselves, but differences between adjacent profile values with respect to the a priori profile are constrained. Since the a priori profile is chosen all zero, this type of regulari-
- sation acts as a smoothing constraint. For methane, above 70 km a diagonal element in the regularisation matrix is used additionally. It pulls the profile in that height region towards the a priori profile. The regularisation matrix entries related to the continuum absorption coefficient contain some off-diagonal elements to prevent neighbouring microwindows from differing too much.
- The retrieval is done on a fine vertical grid with a spacing of 1 km between 4 and 70 km. Above that range the grid is getting coarser and consists of the following grid points: 75, 80, 85, 90, 100 and 120 km.

The retrieval setup version 21 for the full resolution spectra is an adaption of the reduced resolution spectra setup 224 and 225. All changes described in Sect. 3 are

<sup>25</sup> applied to these data versions as well. The only difference is a slightly different selection of the spectral microwindows.



#### 3 Changes in the retrieval setup

As mentioned in the introduction, the main drawback of previous data versions is the high bias of CH<sub>4</sub> and N<sub>2</sub>O below 25 km. In order to improve the data, a number of major modifications in the retrieval setup were adopted. To make our data traceable, all <sup>5</sup> modifications since versions 220 and 221 are reported and discussed in the following.

- The first part (Sects. 3.1 and 3.2) deals with the changes which led to the data product versions 222 and 223. The second part (Sects. 3.3 to 3.5) explains the further changes which were included in the current product versions 224 and 225. In Table 1 the included changes are listed for each data version.
- To illustrate the influence of the individual changes a test dataset of 110 orbits from the reduced resolution measurement period with 10439 limb scans is used. These spectra were measured between 05 June and 18 August 2010. Figure 1 shows the zonal mean vmr distributions for this dataset.

Almost all the changes implemented in the 224 and 225 setup for the reduced resolution spectra have been included in the new 21 setup for the full resolution spectra as well. Only the selection of the spectral microwindows is slightly different. A dataset of 1054 measurements from 16 orbits between 10 January and 20 February 2004 was calculated to investigate this new retrieval setup. In Fig. 2 the latitude dependent mean vmr profiles for those calculations are shown.

#### 20 3.1 Usage of HITRAN 2008

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Previous retrievals of  $CH_4$  and  $N_2O$  (up to versions 220 and 221) relied on the HITRAN 2000 spectroscopic dataset with updates from 2001 for  $N_2O$  and HITRAN 2004 for  $CH_4$ . The spectroscopic datasets of  $CH_4$  and  $N_2O$  received an update in the HITRAN 2008 release (Rothman et al., 2009). Hence in the new  $CH_4$  and  $N_2O$  retrieval setup, these new line datasets have been used.

In Fig. 3 the influence of the usage of the updated spectroscopic dataset is presented. Areas where the new spectroscopy leads to higher mixing ratios are red, those



where it produces lower mixing ratios are blue. Both in  $CH_4$  and  $N_2O$  the mixing ratios are smaller in the lower part of the measured profiles. For  $CH_4$  the main changes are below 20 km in the tropics and below 15 km in the polar regions. The differences are most pronounced in the troposphere at the lower boundary of the data product, where new spectroscopic data lead to values of up to 0.18 ppmv lower than the old ones. These findings agree with Alvarado et al. (2014) who examined the influence of the HITRAN 2008 spectroscopy on the CH<sub>4</sub> profiles retrieved from the NASA AURA Tropospheric Emission Spectrometer (TES, Beer et al., 2001) and found the values to be lower with the new dataset. The N<sub>2</sub>O profiles with the HITRAN 2008 spectroscopy show smaller values up to almost 35 km. Only the lowermost part of the valid data has 10 a difference of less than -10 ppby, the minimum in differences is -19 ppby. The main differences in the retrieved CH<sub>4</sub> and N<sub>2</sub>O result from updates of spectroscopic parameters for lines already existing in the database. Only very small differences between the old and new spectroscopy can possibly be attributed to new lines added within the

used microwindows. 15

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#### 3.2 Spectral offset and continuum contributions

For the data versions 220 and 221 the continuum absorption coefficient was fitted up to an altitude of 32 km, which was the original suggestion of von Clarmann et al. (2003) for the retrieval of temperature. However, in some publications (e.g. Bardeen et al., 2008) relevant aerosol abundances above that altitude are discussed. To account for

contributions from particles in this altitude range, we extended the upper boundary for the fit of the continuum absorption coefficient to 60 km.

The residual radiance offset in versions 220 and 221 has been fitted independently for each individual tangent height. In combination with the fitting of the continuum absorption coefficient, this leads to an ill-posed inverse problem, where the contributions 25 of both these variables can hardly be distinguished: too high continuum absorption coefficients can nearly compensate too low radiance corrections and vice versa. This leads to linearly dependent rows in the Jacobians of the retrieval. Thus, increasing the



altitude range for the fit of the continuum absorption coefficient alone leads to even bigger oscillations. To avoid these instabilities, we now use a constant additive radiative offset over all tangent heights, a hard constraint of the radiance offset which IMK/IAA retrievals of most other species already had applied. This approach avoids any oscillating compensations between spectral offset and the continuum absorption coefficient and leads to smoother profiles of the latter. Convergence does slightly improve. Instead of 10378, now 10403 of a total of 10439 cases do converge. The scientific analysis of the origin of the retrieved background radiation, probably aerosols, remains to be done.

The degrees of freedom (Rodgers, 2000) both in the profiles of methane and nitrous oxide prove to increase slightly. The difference between the retrieval with constant spectral offset and continuum absorption coefficient fitted up to 60 km and the setup versions 220 and 221 is shown in Fig. 4. Both the volume mixing rations of CH<sub>4</sub> and N<sub>2</sub>O are decreased in the altitude range below 20 km. Hence the continuum handling in the old setup led to an effect of downward error propagation and to higher values in these altitude regions. Both gases show an increase at roughly 30 km (in the southern polar regions this feature is at slightly lower altitudes).

#### 3.3 Constraint

The IMK/IAA  $CH_4$  and  $N_2O$  retrievals use an altitude-dependent Tikhonov-like smoothing constraint based on squared first order finite difference matrices  $L_1$ . Up to data <sup>20</sup> versions 222 and 223 the altitude dependence was implemented as suggested by Steck and von Clarmann (2001). This constraint has been replaced by a constraint using a regularisation matrix  $\mathbf{R} = \mathbf{L}_1^T \mathbf{A} \mathbf{L}_1 + \mathbf{D}$  where  $\mathbf{A}$  is a diagonal matrix controlling the altitude dependence of the constraint and  $\mathbf{D}$  is zero except for some diagonal entries above 70 km (increasing with height) affecting methane only. At those altitudes, they pull the profile of methane towards the a priori which is zero. It is introduced be-

cause otherwise the retrieved methane profiles tend to show negative values above 70 km, even in the zonal averages, which clearly is an artefact. Adding the diagonal element in the constraint leads to values close to zero. However, there still remains



some slightly negative retrieved volume mixing ratios in the zonal mean at 75 km. Our analysis shows, that the introduction of the diagonal element to the regularisation matrix **R** does not alter the volume mixing ratio profiles below that altitude. Hence these physically still erroneous values above do not affect the quality of the data product <sup>5</sup> below.

A further change was applied with respect to the off-diagonal matrix elements which pull the continuum absorption coefficient profiles of neighbouring microwindows towards each other. These entries have been considerably reduced, which allows a more pronounced spectral structure of the background emission. This leads to slightly decreased values of  $CH_4$  at 20 km altitude and  $N_2O$  below 15 km. This is shown in Fig. 5, where the difference between the calculations with the new constraint setup minus those with the old setup is plotted. The degrees of freedom of  $CH_4$  were reduced by 0.25 and of  $N_2O$  were increased by 0.25, while the continuum absorption coefficient profiles have gained about 14 degrees of freedom.

#### **3.4 Spectral microwindows**

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Usually high resolution spectroscopy retrievals do not invert the entire measured spectra, but only narrow spectral windows containing lines of the target species, so-called microwindows (e.g. von Clarmann and Echle, 1998). The selection of these spectral microwindows has been changed compared to previous data versions. The microwindows from  $1270 \text{ cm}^{-1}$  towards higher wavenumbers have been restricted to higher altitudes because the spectra in that region were saturated around the position of the line centres below. Additional microwindows at 1225, 1239 and 1245 cm<sup>-1</sup> and in the range of 1257 to 1270 cm<sup>-1</sup> have been introduced to compensate the related loss of information at the higher wavenumbers and to stabilise the joint fit of H<sub>2</sub>O (see Sect. 3.5).

<sup>25</sup> The selection of the microwindows used in versions 220, 221, 222 and 223 are compared to those of versions 224 and 225 in Fig. 6.

Figure 7 shows the impact of the new microwindow selection on the mean profiles of  $CH_4$  and  $N_2O$ . For both species the volume mixing ratios below 20 km did decrease at



all latitudes. In the subtropics and tropics between 20 and 30 km an increase in volume mixing ratio can be observed. Convergence of the iterative retrieval was achieved in a larger part of cases: instead of 10 399 in setup versions 222 and 223, now 10 421 measurements (of 10 439 total) did converge. The changes in the microwindows led to a slight decrease in the root mean square difference between the measured and the

 $_{5}$  a slight decrease in the root mean square difference between the measured and the best-fit spectra (RMS) and to an increase in degrees of freedom for methane. For N<sub>2</sub>O the degrees of freedom dropped slightly.

## 3.5 Joint fit of water vapour and nitric acid

To improve the fit and to reduce the systematic residuals in the best fit spectra, the <sup>10</sup> mixing ratios of HNO<sub>3</sub> and H<sub>2</sub>O are retrieved additionally to CH<sub>4</sub> and N<sub>2</sub>O, continuum absorption coefficient and spectral offset (joint fit approach) in the latest versions. Versions 222 and 223 and earlier used the volume mixing ratio profiles which were retrieved in previous retrieval steps as constant parameters. The new approach can compensate any spectroscopic inconsistency between the spectral microwindows of <sup>15</sup> the specific gas retrievals (in this case HNO<sub>3</sub> and H<sub>2</sub>O) and those used in the setup for the retrieval of CH<sub>4</sub> and N<sub>2</sub>O.

The influence of the joint fit of  $HNO_3$  and  $H_2O$  on the results for  $CH_4$  and  $N_2O$  is shown in Fig. 8. Both the profiles of  $CH_4$  and  $N_2O$  are moderately reduced below 20 km altitude. The RMS does hardly change and the degrees of freedom for  $CH_4$  and  $N_2O$  decrease slightly.

#### 4 Retrieval characterisation

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The  $CH_4$  and  $N_2O$  profiles from the reduced resolution period derived with the new setup versions 224 and 225 show significantly reduced oscillations in polar regions compared to those retrieved with versions 222 and 223. The fraction of converged retrievals of the entire dataset has significantly increased compared to the older versions.



With the new setup 0.37% of the retrievals did not converge, compared to 1.02% of the profiles with setup versions 220 and 221, and 1.27% with versions 222 and 223.

The mean vertical resolution of the test dataset (see Sect. 3) is shown in Fig. 9 both for  $CH_4$  and  $N_2O$ . For  $CH_4$  a good resolution of about 2.5 to 7 km can be obtained below

<sup>5</sup> 60 km. Above it becomes rather coarse, due to the larger tangent height spacing. At altitudes below 20 km there is a slightly degraded resolution. The best resolved part of the profile is the altitude range between 25 and 35 km, where the resolution reaches up to 2.2 km. N<sub>2</sub>O is resolved well between 20 km (15 km in mid latitudes) and 50 km, where the resolution is about 2.5 to 6 km. Above this range the volume mixing ratios
 <sup>10</sup> are too small to allow for a good signal in the spectra. For both gases the mid latitude regions have a better resolution than the tropical or polar areas.

In Fig. 10 the averaging kernels for a sample profile of  $CH_4$  are shown. This reduced resolution scan was measured in orbit 43202 at 39.4° N, 78.9° E on 05 June 2010, 05:02:29 UTC. The upper panel shows the rows of the averaging kernel: each black

- <sup>15</sup> cross on one curve denotes the nominal altitude of the related averaging kernel. The rows of the averaging kernels represent the weights of the true atmospheric states at various altitudes in the retrieval at the nominal altitude. For most curves the black cross lies at the maximum position, i.e., the atmospheric state at a certain altitude has the largest impact on a retrieved value in the same grid height. The curves are
- roughly symmetric to this point in shape. This allows a straight-forward interpretation of the retrieved profiles. Above 55 and below 15 km the curves have their maximum not always on the nominal grid point and/or are asymmetric in shape. Without considering the averaging kernels, this can cause biases, and thus, interpretation of the retrieved profile should be conducted more carefully. The curves in the lower panel picture the
- columns of the averaging kernel. They indicate where the information from a value in the atmosphere (marked with a black cross) impacts the retrieved profile. Again the curves for values between 15 and below 55 km are symmetric and their maxima are located at the retrieved grid points. Figure 11 shows the averaging kernels for the retrieval of N<sub>2</sub>O for the same measurement. Roughly symmetric curves can be found



between 20 and 50 km both in the plots of rows and columns. All the information for the retrieved values above 50 km are almost entirely dependent on the values below. Outside this range the data without explicit consideration of the averaging kernels is prone to misinterpretation.

- <sup>5</sup> Along with the retrieval, for each profile the impact of the instrument noise on the retrieved profile is calculated as a routine data product. However there are various additional error sources which have to be considered. These errors have been estimated for certain example profiles. The following errors have been assumed: for the uncertainty of the line of sight (los) 0.15 km vertical pointing; for the spectral shift 0.005 cm<sup>-1</sup>;
   <sup>10</sup> for the instrumental calibration error (gain) 1 %; for the instrumental line shape error (ils) 3%; and for the temperature gradient in latitudinal direction 0.01 K km<sup>-1</sup> (constant with respect to altitude). The spectroscopic errors were extracted from the HITRAN
  - database (Rothman et al., 2009), with a correction of the actual line intensity in dependence of the rotational quantum number as suggested by Flaud and Piccolo (2001).
- <sup>15</sup> Resulting line intensity uncertainties are between 2 and 5% for  $CH_4$  and between 4 and 7% for  $NO_2$  for low *J* values. The representative uncertainties in air broadening coefficients have been estimated at 15% for  $CH_4$  and 3.5% for  $N_2O$ . Since no information on the error correlations between the individual transitions was available, these errors were assumed to be fully correlated, which implies that the error estimation is
- <sup>20</sup> on the conservative side. The profiles of temperature and  $O_3$  are known from previous retrieval steps; related retrieval errors were propagated onto the CH<sub>4</sub> and N<sub>2</sub>O results. The contribution of CO<sub>2</sub>, SO<sub>2</sub>, NO<sub>2</sub>, HOCI, HCN, H<sub>2</sub>O<sub>2</sub>, C<sub>2</sub>H<sub>2</sub>, COF<sub>2</sub>, CFC-14, N<sub>2</sub>O<sub>5</sub> and CIONO<sub>2</sub> to the spectra was calculated based on climatological abundances. For these gases, estimated profiles of 1-sigma were used to estimate corresponding CH<sub>4</sub>
- and N<sub>2</sub>O retrieval errors. The estimated error contributions of all these sources are shown in Fig. 12 for the measurement at 05 June 2010, 05:02:29 UTC along with the derived profiles. Below 15 km the instrument noise is the most contributing source to the error for both gases. Above, the other errors have larger contributions. Especially the estimated spectroscopic error is very large and dominates the error budget between



about 17 and 42 km for CH<sub>4</sub> and between 20 and 38 km for N<sub>2</sub>O. It has, however, to be mentioned that these spectroscopic error estimates are speculative because the inter-transition correlations of the errors are not known. The assumption of full correlations may be over-conservative. Inter-transition correlations of less than unity would lead to

- <sup>5</sup> partial randomisation of this kind of error and the resulting uncertainty would be largely reduced. The second largest error contribution is the uncertainty of the vertical pointing of the line of sight. For  $CH_4$ , below 60 km the relative total error is between 5 and 17 %. The relative total error of N<sub>2</sub>O below 30 km is around 5 to 10%, above it increases with height to values in the order of 10 to 15%, until it further increases above 40 km.
- <sup>10</sup> Tables 2 and 3 give numeric values for the more important error contributions for a few selected altitude grid points.

For the full resolution spectra, oscillations in the  $CH_4$  profiles were considerably reduced in the version 21. However, a larger fraction of the retrievals did not converge (8.50 % instead of 2.78 %).

Figure 13 shows the mean vertical resolution of a test data set for the full resolution spectra. Between 15 and 40 km the resolution is in the order of 3 to 6 km for both gases. The resolution is not as good as in the period of reduced spectral resolution, because each limb scan consists of fewer tangent altitudes.

The averaging kernels of the test measurement between 15 and 50 km generally look well behaved (Figs. 14 and 15), but in some cases, particularly at very high or low altitudes, are slightly off centre. This should be kept in mind for further interpretation of the data. In most cases the retrieved data points are most sensitive to atmospheric volume mixing ratios slightly below the altitudes they are calculated for (between 1 and 2 km).

The error budget shown in Fig. 16 makes clear that the instrument noise is larger than the other errors at the altitudes below 15 km. Again the largest contributor to the error above that altitude is the spectroscopic error, followed by the uncertainty of the tangent altitude pointing (line of sight). The relative error of  $CH_4$  is between 5 and 20%. For N<sub>2</sub>O it is between 5 and 10% below 25 km and between 5 and 20% between 25



and 45 km. As said before, the spectroscopic error may be over-estimated, because the assumption of inter-transition correlations of these errors may be too pessimistic. Numeric values for certain altitude grid points and the more important error contributions are reported in Tables 4 and 5.

#### 5 5 Conclusions

The new MIPAS-ENVISAT CH<sub>4</sub> and N<sub>2</sub>O profiles versions 21, 224 and 225 are now available for the complete MIPAS measurement period. The usage of the HITRAN 2008 spectroscopic dataset, improved continuum and offset handling, minor changes in the constraint, inclusion of H<sub>2</sub>O and HNO<sub>3</sub> to the retrieval vector and different selection of spectral microwindows overall lead to improved data products where the known high 10 bias has been reduced. Averaging kernels are found to be symmetric in the stratosphere. The vertical resolutions there are in the order of 2.5 to 7 km for  $CH_4$ , and 2.5 to 6 km for N<sub>2</sub>O during the reduced resolution period and in the order of 3 to 6 km for both gases during the full resolution period. The relative errors in the lower part of the profiles are mostly around 15 % for CH<sub>4</sub> and below 10 % for N<sub>2</sub>O. They increase above 25 15 or 30 km to values between 10 and 20 %, except for  $CH_4$  from the reduced resolution period, where the error remains below 15% over almost the entire profile below 60 km. It turned out that knowledge of the air broadening coefficients and line intensities of the individual lines is insufficient to reliably estimate the propagation of spectroscopic

errors on the retrieved vmr profiles. In addition, information on the inter-transition correlations of these errors is needed. Comparisons to other instruments will be the subject of an upcoming paper.

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version names	Temp. coverage	HITRAN 2008	Impr. cont./ons.	new mws	new constraint	Joint fit H <sub>2</sub> O/HNO <sub>3</sub>
V5H_CH4_20	06/2002-03/2004	yes	yes	no	no	no
V5H_N2O_20	06/2002–03/2004	yes	yes	no	no	no
V5H_CH4_21	06/2002-03/2004	yes	yes	yes	yes	yes
V5H_N2O_21	06/2002-03/2004	yes	yes	yes	yes	yes
V5R_CH4_220	01/2005-04/2011	no	no	no	no	no
V5R_N2O_220	01/2005-04/2011	no	no	no	no	no
V5R_CH4_221	05/2011-04/2012	no	no	no	no	no
V5R_N2O_221	05/2011-04/2012	no	no	no	no	no
V5R_CH4_222	01/2005-04/2011	yes	yes	no	no	no
V5R_CH4_222 V5R_N2O_222	01/2005–04/2011 01/2005–04/2011	yes yes	yes yes	no no	no no	no no
V5R_CH4_222 V5R_N2O_222 V5R_CH4_223	01/2005–04/2011 01/2005–04/2011 05/2011–04/2012	yes yes yes	yes yes yes	no no no	no no no	no no no
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V5R_CH4_222 V5R_N2O_222 V5R_CH4_223 V5R_N2O_223 V5R_CH4_224	01/2005-04/2011 01/2005-04/2011 05/2011-04/2012 05/2011-04/2012 01/2005-04/2011	yes yes yes yes	yes yes yes yes	no no no yes	no no no yes	no no no no yes
V5R_CH4_222 V5R_N2O_222 V5R_CH4_223 V5R_N2O_223 V5R_CH4_224 V5R_N2O_224	01/2005-04/2011 01/2005-04/2011 05/2011-04/2012 05/2011-04/2012 01/2005-04/2011 01/2005-04/2011	yes yes yes yes yes	yes yes yes yes yes	no no no yes yes	no no no yes yes	no no no yes yes
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**Table 2.** Error budget for  $CH_4$  for the measurement in orbit 43202 at 39.4° N, 78.9° E on 05 June 2010, 05:02:29 UTC (derived from reduced resolution spectra, version V5R\_CH4\_224). Relative values in %.

Altitude (km)	total	detect noise	line of sight	Т	grad T	ils	gain	spectroscopy	$N_2O_5$	CIONO <sub>2</sub>	Shift
15	5.8	3.1	4.2	0.64	0.38	1.6	1.4	1.2	0.011	0.0025	0.039
20	14.	3.6	3.9	0.80	0.39	1.2	1.5	13.	0.16	0.013	0.22
25	14.	3.4	0.84	0.48	0.27	0.31	2.3	14.	0.18	0.024	0.31
30	12.	2.4	2.0	0.36	0.13	1.5	2.4	11.	0.090	0.11	0.17
40	11.	3.7	0.38	0.38	0.011	5.4	3.5	7.5	0.19	0.16	0.23
50	6.2	3.3	3.0	1.3	0.39	3.1	2.0	1.2	0.056	0.0099	0.089
60	7.9	6.4	3.6	2.1	0.42	0.91	0.68	0.37	0.0087	0.0020	0.087

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Table 3. As Table 2, but for  $N_2O$  (version V5R\_N2O\_224).

Altitude (km)	total	detect noise	line of sight	Т	grad T	ils	gain	spectroscopy	$N_2O_5$	CIONO <sub>2</sub>	Shift
15	5.9	3.1	3.7	0.56	0.31	1.4	1.1	2.7	0.074	0.0040	0.021
20	8.6	2.5	4.7	0.94	0.43	0.94	1.5	6.3	0.13	0.0039	0.016
25	7.8	2.3	0.62	0.56	0.29	1.7	2.9	6.7	0.15	0.030	0.16
30	6.5	1.8	3.0	0.46	0.080	2.0	1.9	4.7	0.071	0.11	0.0071
40	17.	6.8	5.0	1.3	0.16	10.	6.6	9.6	0.74	0.31	0.20
50	10.	8.3	3.3	0.64	0.18	3.1	1.8	3.9	0.045	0.0031	0.25

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**Table 4.** Error budget for  $CH_4$  for the measurement of in orbit 10324 at 46.0° N, 144.5° W on 20 February 2004, 07:48:31 UTC (derived from full resolution spectra, version V5H\_CH4\_21). Relative values in %.

Altitude (km)	total	detect noise	line of sight	Т	grad T	ils	gain	spectroscopy	$N_2O_5$	CIONO <sub>2</sub>	Shift
15	11.	2.5	3.9	1.3	0.63	0.13	0.51	9.7	0.21	0.027	0.22
20	13.	2.5	4.1	0.75	0.36	0.22	0.71	13.	0.11	0.011	0.16
25	15.	2.4	5.4	0.92	0.33	0.077	0.22	14.	0.15	0.048	0.27
30	20.	4.5	5.4	0.38	0.35	0.71	0.92	18.	0.14	0.074	0.93
40	11.	8.5	1.6	1.1	1.5	6.0	3.3	0.12	0.23	0.066	1.1
50	17.	8.1	1.9	0.44	1.5	8.1	3.5	11.	0.13	0.0040	0.74
60	17.	15.	4.8	3.5	0.070	1.5	1.2	1.2	0.0011	0.0056	0.35

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Table 5. As Table 4, but for  $N_2O$  (version V5H\_N2O\_21).

Altitude (km)	total	detect noise	line of sight	Т	grad T	ils	gain	spectroscopy	$N_2O_5$	CIONO <sub>2</sub>	Shift
15	8.2	1.9	4.8	1.6	0.79	0.17	0.65	6.2	0.16	0.058	0.13
20	10.	2.0	5.4	1.0	0.41	0.11	0.77	8.2	0.082	0.011	0.020
25	8.2	1.8	5.6	0.97	0.51	0.16	0.16	6.1	0.082	0.0072	0.17
30	24.	7.6	16.	0.56	0.80	3.5	1.2	16.	0.068	0.032	0.056
40	13.	7.2	0.085	0.24	1.2	6.6	3.9	7.2	0.048	0.0085	0.60
50	39.	34.	12.	0.94	1.1	6.8	3.3	10.	0.11	0.059	0.33





**Figure 1.** Zonal mean distributions for reduced resolution spectra, measured between 05 June and 18 August 2010. The black lines are the interpolated isopleths. White areas show regions with no valid data at all. On the upper panel for  $CH_4$  (version V5R\_CH4\_224), on the lower for N<sub>2</sub>O (version V5R\_N2O\_224).















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Figure 5. As Fig. 3, but difference retrieval with Tikhonov constraint and relaxed constraint between neighbouring microwindows minus reference with old setup.





**Figure 6.** Selected microwindows of the setups V5R\_CH4\_220, V5R\_CH4\_221, V5R\_N2O\_220 and V5R\_N2O\_221 as well as V5R\_CH4\_222, V5R\_CH4\_223, V5R\_N2O\_222 and V5R\_N2O\_223 (upper panel) and V5R\_CH4\_224, V5R\_CH4\_225, V5R\_N2O\_224 and V5R\_N2O\_225 (lower panel) as a function of wavenumbers and tangent altitudes. The microwindows are marked as black patches.





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**Figure 9.** Mean vertical resolution in km for profiles from reduced resolution spectra (versions V5R\_CH4\_224 and V5R\_N2O\_224). Upper panel for  $CH_4$ , lower panel for  $N_2O$ .





**Figure 10.** Selected rows (upper panel) and columns (lower panel) of the averaging kernel matrix for the measurement of  $CH_4$  in orbit 43202 at 39.4° N, 78.9° E on 05 June 2010, 05:02:29 UTC (derived from reduced resolution spectra, version V5R\_CH4\_224). The black crosses highlight the diagonal terms of the averaging kernel matrix.





Figure 11. As Fig. 10, but for N<sub>2</sub>O (version V5R\_N2O\_224).





**Figure 12.** Estimated error contributions for the measurement in orbit 43202 at  $39.4^{\circ}$  N, 78.9° E on 05 June 2010, 05:02:29 UTC (derived from reduced resolution spectra, versions V5R\_CH4\_224 and V5R\_N2O\_224). Left panels: gas profiles, right panels: contributions of different errors. All contributions are absolute values, except for the total relative error, which is given in % (upper axis). Upper panels for CH<sub>4</sub>, lower panels for N<sub>2</sub>O.







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**Figure 14.** Selected rows (upper panel) and columns (lower panel) of the averaging kernel matrix for the measurement of  $CH_4$  in orbit 10324 at 46.0° N, 144.5° W on 20 February 2004, 07:48:31 UTC (derived from full resolution spectra, version V5H\_CH4\_21). The black crosses highlight the diagonal terms of the averaging kernel.





Figure 15. As Fig. 14, but for N<sub>2</sub>O (version V5H\_N2O\_21).





**Figure 16.** As Fig. 12, but for the measurement of in orbit 10324 at 46.0° N, 144.5° W on 20 February 2004, 07:48:31 UTC (version V5H\_N2O\_21).

