**Atmospheric
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Remote sensing of CO_2 and CH_4 using solar absorption spectrometry with a commercial low resolution spectrometer

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Abstract

Throughout the last few years solar absorption Fourier Transform Spectrometry (FTS) has been further developed to measure the total columns of CO_2 and CH_4. The observations are performed at high spectral resolution, typically at 0.02 cm^{-1}. The precision achieved is actually generally better than 0.25 %. However, these high resolution instruments are quite large and need a dedicated room or container for installation. We performed these observations using a smaller commercial interferometer at its maximum possible resolution of 0.11 cm^{-1}. The measurements have been performed at Bremen and have been compared to observations using our high resolution instrument also situated at the same location. The high resolution instrument has been successfully operated as part of the Total Carbon Column Observing Network (TCCON). The precision of the low resolution instrument is 0.32 % for XCO_2 and 0.46 % for XCH_4. A comparison of the measurements of both instruments yields an average deviation in the retrieved daily means of ≤0.2 % for CO_2. For CH_4 an average bias between the instruments of 0.46 % was observed. For test cases, spectra recorded by the high resolution instrument have been truncated to the resolution of 0.11 cm^{-1}. This study gives an offset of 0.03 % for CO_2 and 0.26 % for CH_4. These results indicate that for CH_4 more than 50 % of the difference between the instruments results from the resolution dependant retrieval. We tentatively assign the offset to an incorrect a-priori concentration profile or the effect of interfering gases, which may not be treated correctly.

1 Introduction

The investigation of the sources and sinks of greenhouse gases requires accurate measurements of their atmospheric concentrations. So far, knowledge on the atmospheric burden of CO_2 and CH_4 is mainly based on in-situ measurements, which sample the air at the surface. In addition, sporadic aircraft observations in the lower atmosphere and a few tall tower measurements are available. However, using surface data in inverse

models requires assumptions on the vertical mixing of the air-masses (volume mixing ratio, VMR). Total column measurements on the other hand are much less influenced by vertical mixing, which reduces the assumptions made in inverse models (Yang et al., 2007).

Remote sensing has been established as a powerful tool in atmospheric science. Using the sun or moon as light source, up to 30 trace gases can be observed in the infrared spectral region. These observations yield, first of all, the total columns of these trace gases. These total column measurements can be performed either from the ground by upwards-looking solar absorption spectrometry, or by downward-looking satellites using sunlight, reflected at the earth surface. Over the last few years ground-based solar absorption Fourier Transform Spectrometry (FTS) and especially the retrieval algorithm has been further developed to measure the averaged total column mixing ratio of CO_2 and CH_4 with high precision. In order to minimize systematic errors and obtain the required precision, the column averaged dry-air mole fractions of CO_2, named XCO_2, are determined by normalizing them to the known dry-air mole fractions of O_2 of 20.95 %.

$$XCO_2\ (O_2) = 0.2095 * \frac{\text{column } CO_2}{\text{column } O_2}. \tag{1}$$

and

$$XCH_4\ (O_2) = 0.2095 * \frac{\text{column } CH_4}{\text{column } O_2}. \tag{2}$$

This reduces many of the systematic errors, as both VMRs are achieved in the same way.

These ground-based observations are organized into a global measurement community, the Total Carbon Column Observatory Network TCCON (Wunch et al., 2011). Within TCCON, the high resolution Bruker interferometers (IFS 120 HR and 125 HR) have been widely accepted as preferred instruments as they have demonstrated the long and short-term stability required by TCCON standards. This is possible due to

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their large light throughput, coupled with high spectral resolution, thus achieving sufficient precision and accuracy. Spectra are taken at maximum optical path differences between 45 cm and 65 cm, corresponding to resolutions varying between 0.014 cm^{-1} and 0.02 cm^{-1} respectively, where the resolution is defined (following the Bruker notation) as resolution = 0.9/OPD, where the OPD is the maximum optical path difference.

XCO_2 can now be measured with a precision of better than 0.25 % and XCH_4 to better than 0.4 % (Yang et al., 2002; Warneke et al., 2005; Washenfelder et al., 2006; Deutscher et al., 2010; Messerschmidt et al., 2010; Wunch et al., 2010). The accuracy has been determined by aircraft campaigns to be in the order of 0.2 % for XCO_2 an 0.4 % for XCH_4 (Deutscher et al., 2010; Messerschmidt et al., 2010; Wunch et al., 2010). During these calibration campaigns aircraft capable of flying to high altitudes equipped with in-situ samplers have been used to create total columns of CO_2 and CH_4. All European TCCON instruments have been calibrated in 2009 within the EU project IMECC (Messerschmidt et al., 2011).

TCCON-instruments are quite large and require a separate container or specific laboratory. It is therefore desirable to use a more compact instrument. Low resolution instruments require much less space (their size being dictated largely by the maximum OPD) and can easily be used in remote areas or for short campaigns. Besides working as a travel standard, low resolution measurements are of interest in understanding differences between the TCCON observations and satellite measurements (which are recorded at low resolution). The resolutions of the three dedicated satellite greenhouse gas instruments SCIAMACHY, TANSO and OCO-2 are 7.0 cm^{-1}, 0.2 cm^{-1} and 0.3 cm^{-1} respectively. Since the individual spectral lines are not fully resolved at these resolutions in the spectra recorded by these satellites, the retrieval requires a good knowledge of the background intensity and the underlying interfering gases.

Before using low resolution ground-based instruments in any network as an operational instrument, a validation and calibration procedure is required. The obvious choice to perform such a validation/calibration campaign is to use high resolution TCCON instruments as a reference. Since our high resolution IFS 125 HR has been

calibrated within the IMECC aircraft campaign it can serve as appropriate reference for the IFS 66. In this paper we present ground-based solar absorption observations of CO$_2$ and CH$_4$ using the small commercial interferometer Bruker IFS 66 at its maximum possible resolution of 0.11 cm^{-1}. The measurements have been performed in Bremen/Germany (53.1° N, 8.8° E) for nine days between winter 2009 and spring 2010.
5 The results have been compared to observations performed by our high resolution TCCON instrument running at the same site in Bremen. Besides the direct comparison, the effect of the resolution on the retrieved columns has been investigated by truncating the high resolution spectra to a range of different resolutions between 0.014 cm^{-1}
10 and 0.5 cm^{-1}.

2 Instrumentation

The IFS 66 was installed in a room at our institute above the laboratory where the IFS 125 HR is housed. A home-made solar tracker, driven by a quadrant diode, is installed on the top of the building, feeding a parallel light beam onto the entrance aper-
15 ture of the IFS 125 HR. In order to minimize the influence of the solar tracker (i.e. pointing errors), the same solar tracker as used by the high resolution instrument was used by the IFS 66. We inserted a flat mirror at 45° into the vertical light beam to direct the sunlight from the solar tracker to the IFS 66, intercepting the sunlight to the high resolution instrument. This allows us to alternate measurements between the instruments.
20 Spectra were recorded with the IFS 66 at a maximum resolution of 0.11 cm^{-1} and with the IFS 125 HR at 0.014 cm^{-1}. In the IFS 125 HR the moving mirror is driven on a set of polished rails while the IFS 66 uses a frictionless air bearing. Both instruments use filter wheels to select the aperture. The aperture of the IFS 125 HR was set to 1.0 mm diameter, corresponding to a field of view of 0.0024 radians. For the IFS 66 we used
25 an aperture of diameter 0.25 mm, resulting in a field of view of 0.0016 radians. While the diameter of the parallel light beam of the IFS 125 HR is 6.5 cm, the parallel beam of the IFS 66 is only 3.5 cm. Both instruments were equipped with CaF$_2$ beamsplitters

and indium gallium arsenide (InGaAs) IR-detectors, working at room temperature. Furthermore, both instruments were purged with dry air.

3 Measurements and analysis

Measurements from both instruments were recorded between November 2009 and
5 April 2010 on nine clear days. The resulting solar absorption spectra were obtained alternately by changing the optical path of the sunlight between the instruments after each 30 min period. For the IFS 125 HR two scans were averaged, while ten scans were averaged for the IFS 66. This leads to comparable observation times for each spectrum. For both instruments we used the commercial OPUS software package
10 supplied by Bruker to record and transform the interferograms into spectra for later processing. To reduce the impact of source brightness fluctuations due to changes in the atmosphere, the retrieval strategy within TCCON is to normalize the DC recorded interferograms with the low-pass filtered and smoothed signal (Keppel-Aleks, 2007). We did not apply this DC correction as the IFS 66 is not able to record a DC signal.
15   The analysis was performed using the least-squares algorithm GFIT, developed at NASA/JPL (Toon et al., 1992). The algorithm has been adapted for TCCON (Wunch et al., 2011). GFIT scales an assumed a-priori concentration profile until the simulated spectra best fits the observations by minimizing the RMS residual. In our analysis we used a beta-version of GFIT, that has been modified to pre-adjust the stratospheric
20 a-priori concentration of CH_4 by making use of the known correlation between stratospheric CH_4 and HF. In the stratosphere HF is a very stable trace gas. The tropospheric burden of HF can be neglected compared to the stratospheric one. Therefore the total columns of HF, representing only the stratospheric burden, allow building a more realistic stratospheric a-priori profile of CH_4 (Washenfelder et al., 2003). This leads to
25 better spectral fits for CH_4, as discussed later.

4 Results

4.1 Alignment

The alignment of both instruments is checked by cell measurements using an HCl gas-cell of known pressure and temperature. The instrumental line shape (ILS) was retrieved with the "LINEFIT" program using HCl lines from 5680 to 5800 cm^{-1} (Hase, 1999). The IFS 125 HR typically gives a variation in the modulation efficiency of ≤5 % over the full OPD. For the IFS 66 the modulation efficiency that has been measured directly after an alignment, increases linearly from 1.0 to 1.05 over the optical path difference of 8.1 cm. For this kind of optical set up, using a 90° off-axis parabolic mirror in the interferometer, along with a frictionless air bearing for the movable mirror, a 5 % change in the modulation efficiency over the whole optical path length is acceptable (Bruker, personal communication). However, the alignment of the IFS 66 was found to be much more temperature sensitive than for the IFS 125 HR. Repeating the ILS measurements for conditions where the temperature of the whole laboratory was warmer by 10 °C resulted in an increase in the modulation efficiency of 2 % (see Sect. 4.5 below).

As an example, Fig. 1 shows part of the spectral fit in one CH_4 absorption window. Figure 1a shows the measurements by the IFS 125 HR at a resolution of 0.014 cm^{-1} (64.3 cm OPD). Figure 1b shows the same spectrum as in Fig. 1a, but the interferogram has been truncated to 0.11 cm^{-1} (8.1 cm OPD), corresponding to the resolution of the IFS 66. Figure 1c shows the spectrum measured by the IFS 66 at a resolution of 0.11 cm^{-1} (8.1 cm). The residuals of Fig. 1c have systematic deviations at the absorption lines, which we assign to non-optimal alignment of the interferometer.

4.2 Precision and comparability

Results for individual measurements of XCO_2 and XCH_4 are shown in Figs. 2 and 3. The error bars indicate the typical precision (one sigma), which was determined by looking at the variability within one day. For the IFS 66 we obtained a precision of

0.32 % (1.2 ppm) for CO$_2$ and 0.41 % for CH$_4$ (7.3 ppb). For comparison, the network wide precision of the IFS 125 HR was found to be 0.25 % (1.0 ppm) for CO$_2$ and 0.22 % (3.9 ppb) for CH$_4$, in agreement with (Wunch et al., 2010, 2011). It is important to note that these results were achieved for comparable observation times, two scans for the IFS 125 HR and ten scans for the IFS 66. Tables 1 and 2 give individual results for the nine days on which comparability measurements were possible. In addition to the derived precision the uncertainty based on the spectral fit and residuals is given.

Comparing the results from both instruments gives an average deviation of 0.09 % or 0.35 ppm for XCO$_2$ which is within the precision of both instruments (Fig. 4). Results for XCH$_4$ are shown in Fig. 5. For XCH$_4$ the results from the IFS 66 are lower by 0.46 % compared to the IFS 125 HR. The offset is constant, and larger than the precision. After subtracting this offset the average deviations of both instruments is 0.15 %, in agreement with the precision of both instruments.

Before the 22 January 2010, the IFS 66 was not well aligned, so the effect of an improperly aligned instrument on the resulting XCO$_2$ and XCH$_4$ could be tested by comparing the results.

While XCO$_2$ is only slightly affected by a bad alignment, the deviations for XCH$_4$ increase by a factor of three, as given in Tables 1 and 2.

4.3 Signal-to-noise ratio

The measurements for the IFS 66 show a larger scatter and lower precision compared to the IFS 125 HR, although the measurement times are comparable. We assign this larger scatter to the different light throughput in the two instruments, which results from the differences in the beam diameter and field of view. The difference in the parallel light beam diameter results in an intensity ratio of

$$\text{Int}_{\text{diam(125)}}/\text{Int}_{\text{diam(66)}} = 6.5^2/3.5^2 = 3.5. \tag{3}$$

The difference in the field of view gives a ratio of

$$\text{Int}_{\text{fv(125)}}/\text{Int}_{\text{fv(66)}} = 1.3. \tag{4}$$

The overall ratio of

$$\text{Int}_{125}/\text{Int}_{66} = 4.6 \tag{5}$$

converts to ratio for the Signal-to-noise ratio (S/N) of

$$\sqrt{4.6} = 2.1. \tag{6}$$

5 The measurements give a ratio between 2 and 4, in reasonable agreement with our estimations.

4.4 Influence of resolution

When comparing XCH_4 obtained from both instruments, we attribute the offset of 0.46 % to a combination of improper a-priori profiles, erroneous spectroscopic data,
10 and / or incorrect assumptions in the pT-profile in the retrieval. The residuals in Fig. 1 indicate a non-perfect fit. We speculate that non-ideal choice of input parameters for the least-squares fitting procedure have different effects for the low resolution spectra compared to the high resolution ones. In order to test the effect of the resolution, we truncated the high resolution interferograms of the IFS 125 HR to a resolution of
15 $0.11 \, \text{cm}^{-1}$, the resolution used by the IFS 66. The results are shown in Figs. 6 and 7. While XCO_2 shows almost no dependence on the resolution (0.02 %), for XCH_4 a constant offset of 0.26 % can be observed, in agreement with our estimation given above.

In order to investigate this in more detail, we truncated the high resolution in-
20 terferograms of the IFS 125 HR in several steps down to a resolution of $0.5 \, \text{cm}^{-1}$, which is comparable to the satellite spectrometers SCIAMACHY / ENVISAT and TANSO / GOSAT. The results are shown in Figs. 8a–c and 9a–c. The tests have been performed for all spectra measured on 4 September 2009. Shown is the daily mean. For reference, the variability in percent for each compound is given as a bar in the fig-
25 ure. The dependence on the OPD increases with decreasing OPD. Down to an OPD of 6 cm ($0.15 \, \text{cm}^{-1}$), the O_2 and CO_2 columns show only a weak dependency of 0.02 %.

For CH_4 columns, the variation is much larger, between 0.05 % and 0.3 %, depending on the spectral band analyzed. The corresponding results for XCO_2, XCH_4 and the most important interfering gas, H_2O, are also shown. The dependency for H_2O is about 0.1 %. As there are H_2O lines in the window where CH_4 is retrieved (5938 cm^{-1},

5   6002 cm^{-1} and 6076 cm^{-1}), for low resolution spectra the lines cannot be separated. It could be expected that the retrieval for low resolution spectra interprets H_2O absorption as CH_4 absorption and the retrieved values for these gases would be anti-correlated with a decreasing resolution. Interestingly, that is not the case.

These simulations for XCO_2 and XCH_4 are in agreement with the results from the

10   comparison of both instruments, as shown in Figs. 2 and 3. While the CO_2 total columns for both instruments agree within the precision, for CH_4, a constant bias of 0.26 % was found.

In order to investigate the dependency of CH_4 on the OPD in more detail, we shifted the whole a-priori VMR profile of CH_4 down by 4 km, and repeated the resolution-

15   dependant sensitivity study shown in Figs. 8c and 9c. The results are displayed in Figs. 8d and 9d. The simulations show that the dependency of the resolution on the CH_4 total column is less. Furthermore, the dependency of the windows at 6002 cm^{-1} and 6076 cm^{-1} nearly compensate each other. We tried also other vertical shift amounts, i.e. 2, 4, 6 and 8 km, with 4 km yielding the "optimum" result. That is,

20   4 km yielded the smallest dependency on the OPD. This might indicate that the a-priori and/or the spectral data of CH_4 are erroneous. For completeness, we have repeated the simulations for CO_2 and O_2 but the results do not depend on shifting the a-priori up and down (not shown), which is not surprising, as the volume mixing ratio of both trace gases show only weak variability with altitude.

25   Our study indicates that the retrieved columns depend on the resolution, especially if the assumed a-priori VMR profile is not perfect. However, it must be mentioned that within TCCON, the retrieval strategy applied so far is profile scaling. When applying a complete profile retrieval algorithm the OPD dependency will look different. Differences for CH_4 total columns depending on the retrieval algorithm have already been observed

by Petersen et al. (2010), who compared profile scaling with the optimal estimation approach for tropical spectra from Suriname.

For an OPD of less than $6\,cm$ ($0.15\,cm^{-1}$) the dependency of the total columns increases for decreasing OPD. For such a low resolution, the individual lines are not resolved and we speculate that the influence of the curvature of the whole spectral region analyzed begins to dominate the fitting. The curvature is due to the wavelength dependent sensitivity of the detector, the reflectivity of all mirrors, the filters applied, and the spectral wings of interfering gases. This dependency can partly be avoided by additional measurements using a blackbody radiation light source to obtain an absolute calibration of the spectra. However, this is beyond the scope of this paper and will be investigated in a separate study.

4.5 Stability

As mentioned previously, the ILS of the instrument was found to be dependent on the temperature of the instrument. The comparison for XCO_2 by the IFS 66 with the IFS 125 HR indicates good agreement between both instruments, independent of the alignment of the IFS 66. In comparison, the results for XCH_4 depend on the alignment, see Tables 1 and 2. Tentatively, we assign this dependency to the combination of the least-squares fitting together with an imperfect a-priori VMR. When using a suitable a-priori, the deviations resulting from a misaligned instrument partly compensate, but for an a-priori that is far from ideal the residuals of the fit, combined with the quadratic dependency of the least-squares method, yield shifts in the total columns.

5 Summary and conclusion

The precision for the low resolution instrument was found to be 0.32 % for XCO_2 (1.2 ppm) and 0.41 % (7.3 ppb) for XCH_4. For comparable measurement times the precision of the IFS 66 is lower compared to the IFS 125 HR, which can be explained

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by the lower light throughput of the IFS 66. The comparison of both instruments gives good agreement, deviation of daily means is below 0.2 % (0.6 ppm) for XCO$_2$. For XCH$_4$ a constant offset of 0.46 % has been observed. After subtracting the constant offset, the average deviation between the instruments was also of the order of 0.2 %

5 (4 ppb). We assign the offset to errors in the a-priori profile and or erroneous spectroscopic data for CH$_4$. Truncating the high resolution interferograms of the IFS 125 HR so the resolution is equivalent to the resolution of the IFS 66 gives an offset of 0.26 %, which partly explains the measured offset of the IFS 66 for CH$_4$. The ILS of the low resolution instrument was found to depend on the temperature. While the results for

10 XCO$_2$ do not depend on the temperature and alignment, for XCH$_4$ the precision and comparability require a well aligned instrument. Overall we can conclude that XCO$_2$ can be measured with a low resolution FTS instrument with sufficient precision and accuracy, however, for XCH$_4$ the results depend on the alignment.

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Table 1. Daily means of XCO$_2$ (O$_2$) for nine measuring days.

Day	XCO$_2$ (O$_2$) IFS-125 HR [ppmV]	1σ [ppmV]	XCO$_2$ (O$_2$) IFS-66 [ppmV]	1σ [ppmV]	Difference [%]
20 November 2009	385,00	0,48	385,27	1,05	0,07
1 December 2009	387,53	0,25	387,56	1,13	0,01
28 December 2009	388,59	2,98	388,55	1,90	0,01
22 January 2010	389,07	0,60	388,36	0,76	0,18
4 March 2010	392,20	2,17	392,55	1,63	0,09
5 March 2010	392,02	0,18	391,63	1,17	0,10
9 March 2010	392,44	1,61	392,69	1,38	0,06
12 April 2010	391,87	0,32	391,32	0,90	0,14
15 April 2010	393,00	0,25	392,48	1,17	0,13
average	390,19	0,98	390,05	1,23	0,09

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Table 2. Daily means of XCH$_4$ (O$_2$) for nine measuring days.

Day	XCH$_4$ (O$_2$) IFS-125 HR [ppmV]	σ [*10^{-2} ppmV]	XCH$_4$ (O$_2$) IFS-66 [ppmV]	σ [*10^{-2} ppmV]	Difference [%]	Diff. –offset [%]
20 November 2009	1,776	0,52	1,746	1,29	1,70	2,15
1 December 2009	1,785	0,17	1,771	1,14	0,75	1,21
28 December 2009	1,798	0,73	1,770	0,53	1,55	2,00
22 January 2010	1,803	0,45	1,792	0,57	0,61	0,87
average before alignment	1,791	0,47	1,770	0,88	1,15	1,41
4 March 2010	1,777	1,11	1,788	0,79	0,65	0,19
5 March 2010	1,785	0,18	1,796	0,79	0,59	0,13
9 March 2010	1,795	0,39	1,801	0,71	0,33	0,13
12 April 2010	1,768	0,20	1,773	0,83	0,23	0,23
15 April 2010	1,794	0,07	1,803	0,51	0,51	0,05
average after alignment	1,784	0,39	1,792	0,73	0,46	0,15

Fig. 1. Small part of a measured and simulated spectrum in the region of CH_4. The upper left Figure shows the high resolution spectrum of the IFS 125 HR at a resolution of $0.014\,\text{cm}^{-1}$ (OPD = 64 cm), the upper right one is based on the same measurement, but the interferogram has been truncated to a resolution of $0.11\,\text{cm}^{-1}$ (OPD = 8.1 cm), and the lower one shows a spectrum measured by the IFS 66. at a resolution of $0.11\,\text{cm}^{-1}$.
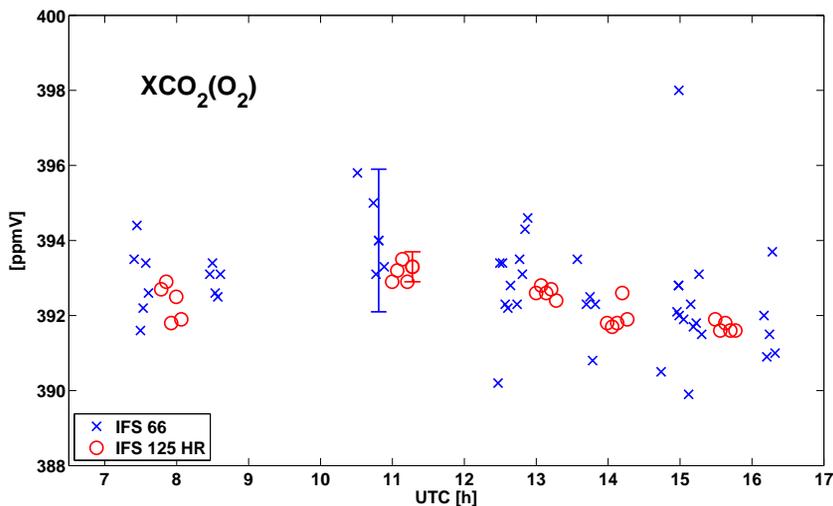
Fig. 2. Individual results for the average mixing ratio of CO$_2$, as measured by both instruments for the 9 March 2010. 10 interferograms have been averaged for the IFS 66 for one data point, and 2 interferograms for the IFS 125 HR. With typical errorbars for single measurements.
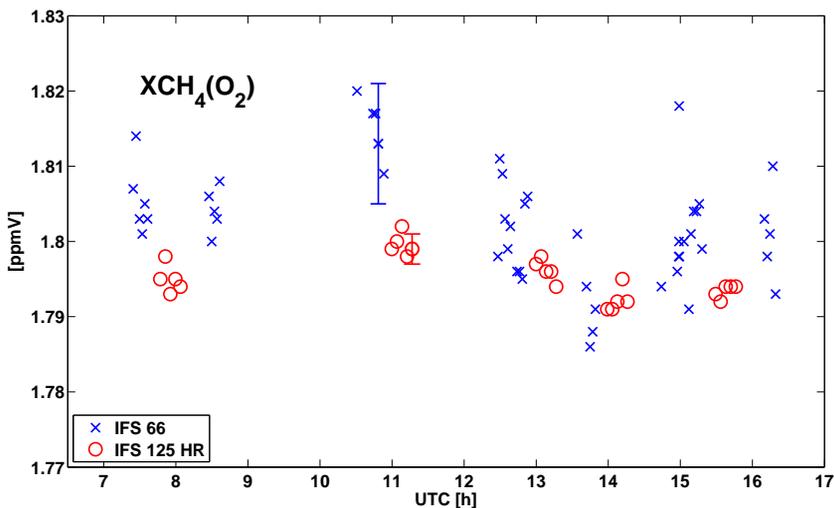
Fig. 3. Individual results for the average mixing ratio of CH$_4$, as measured by both instruments for the 9 March 2010. 10 interferograms have been averaged for the IFS 66 for one data point, and 2 interferograms for the IFS 125 HR. With typical errorbars for single measurements.
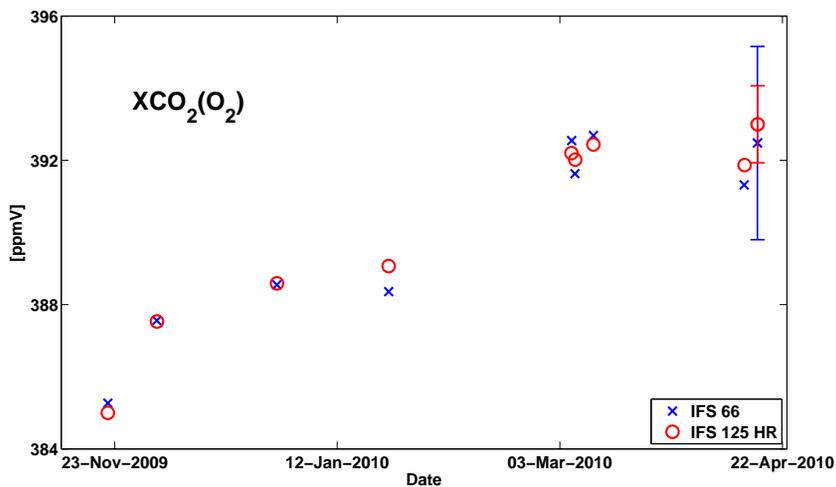
Fig. 4. Daily averages of XCO$_2$, as measured by the IFS 66 and the IFS 125 HR. With typical errorbars.
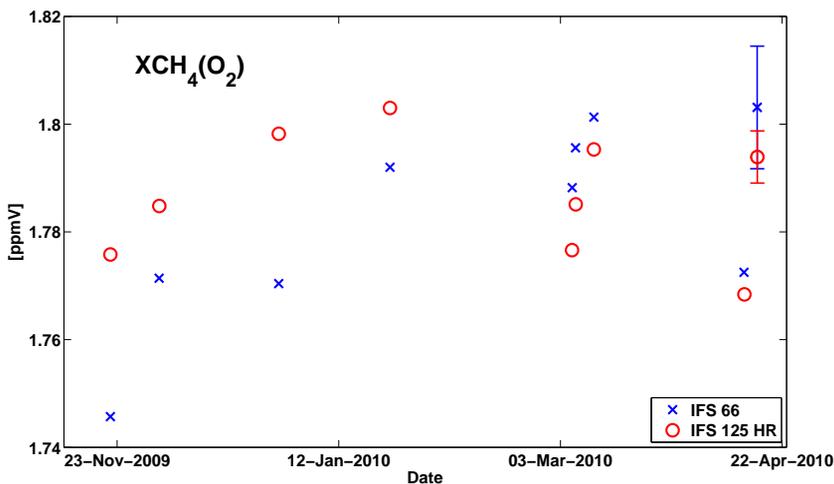
Fig. 5. Daily averages of XCH$_4$, as measured by the IFS 66 and the IFS 125 HR. With typical errorbars.
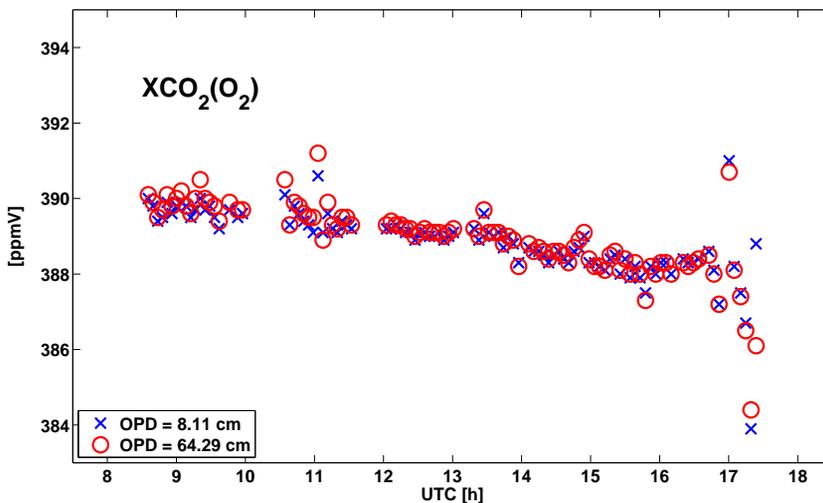
Fig. 6. Retrieved XCO$_2$ from observations by the high resolution instrument IFS 125 HR for the 4 April 2009. The red symbols correspond to the high resolution of 0.014 cm^{-1}, the blue symbols to a resolution of 0.11 cm^{-1}.

XCH$_4$(O$_2$)

[ppmV]

OPD = 8.11 cm
OPD = 64.29 cm

UTC [h]
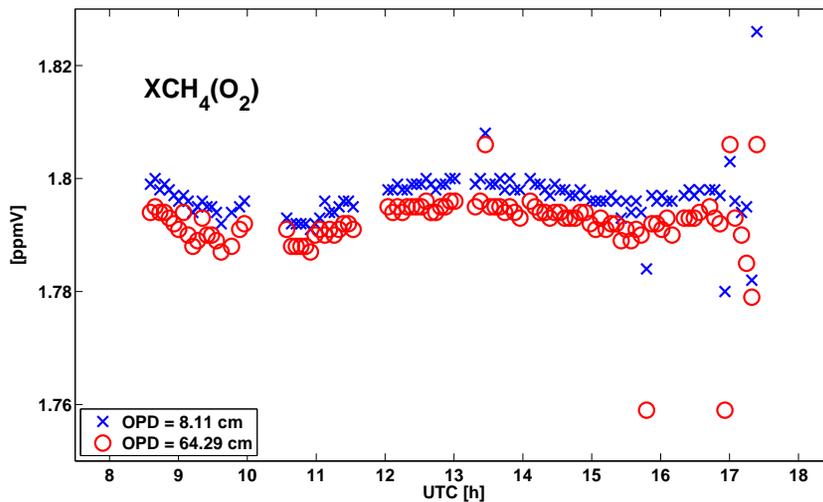
Fig. 7. Retrieved XCH$_4$ from observations by the high resolution instrument IFS 125 HR for the 4 April 2009. The red symbols correspond to the high resolution of 0.014 cm^{-1}, the blue symbols to a resolution of 0.11 cm^{-1}.
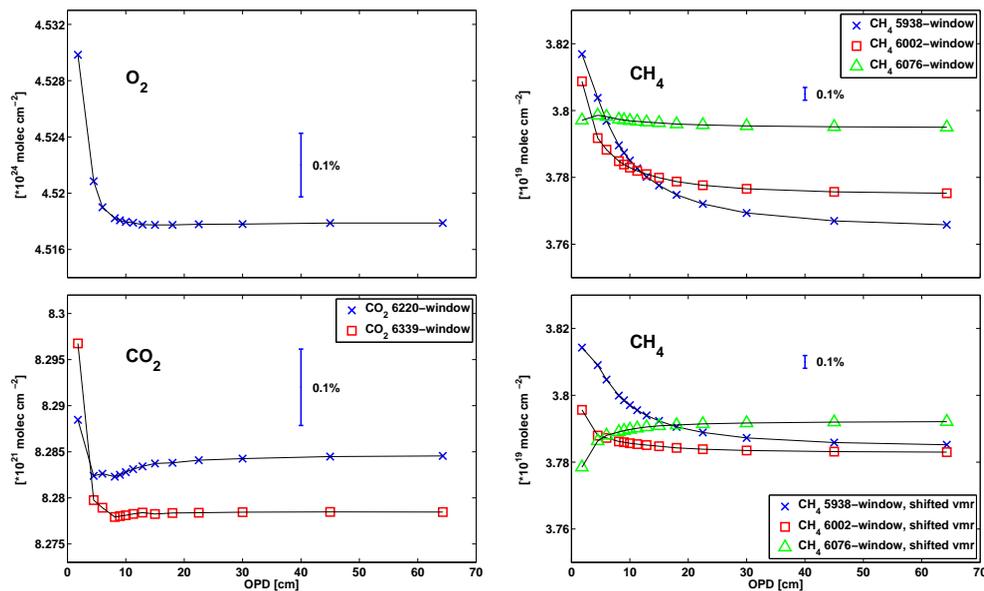
Fig. 8. Retrieved O$_2$, CO$_2$ and CH$_4$ total columns from the IFS 125 HR, by truncating the interferograms to different optical path differences. For CO$_2$ and CH$_4$ the different retrieval windows are shown. For CH$_4$ the initial a-priori profile has been shifted vertically by −4 km.
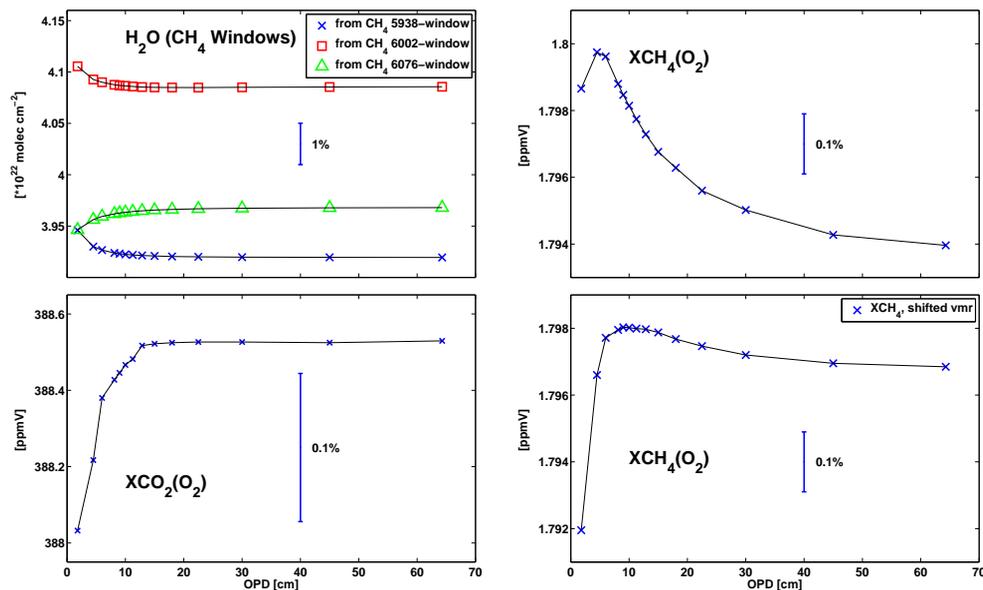
Fig. 9. Retrieved H$_2$O, XCO$_2$ and XCH$_4$ from the IFS 125 HR, H$_2$O is taken from the different retrieval windows of CH$_4$. For H$_2$O the different retrieval windows are shown. For XCH$_4$ the initial a-priori profile has been shifted vertically by −4 km