



# Remote sensing of CO<sub>2</sub> and CH<sub>4</sub> using solar absorption spectrometry with a low resolution spectrometer

C. Petri<sup>1</sup>, T. Warneke<sup>1</sup>, N. Jones<sup>2</sup>, T. Ridder<sup>1</sup>, J. Messerschmidt<sup>1</sup>, T. Weinzierl<sup>1</sup>, M. Geibel<sup>3,\*</sup>, and J. Notholt<sup>1</sup>

<sup>1</sup>IUP, University of Bremen, Bremen, Germany

<sup>2</sup>School of Chemistry, University of Wollongong, Wollongong, Australia

<sup>3</sup>Max Planck Institute for Biogeochemistry, Jena, Germany

\* now at: Department for Applied Environmental Research (ITM), Stockholm University, Stockholm, Sweden

Correspondence to: C. Petri (christof.petri@iup.physik.uni-bremen.de)

Received: 7 November 2011 – Published in Atmos. Meas. Tech. Discuss.: 9 January 2012

Revised: 18 May 2012 – Accepted: 3 June 2012 – Published: 12 July 2012

**Abstract.** Throughout the last few years solar absorption Fourier Transform Spectrometry (FTS) has been further developed to measure the total columns of CO<sub>2</sub> and CH<sub>4</sub>. The observations are performed at high spectral resolution, typically at 0.02 cm<sup>-1</sup>. The precision currently achieved is 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<sup>-1</sup>. 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<sub>2</sub> and 0.46 % for XCH<sub>4</sub>. A comparison of the measurements of both instruments yields an average deviation in the retrieved daily means of ≤0.2 % for CO<sub>2</sub>. For CH<sub>4</sub> an average bias between the instruments of 0.47 % was observed. For test cases, spectra recorded by the high resolution instrument have been truncated to the resolution of 0.11 cm<sup>-1</sup>. This study gives an offset of 0.03 % for CO<sub>2</sub> and 0.26 % for CH<sub>4</sub>. These results indicate that for CH<sub>4</sub> more than 50 % of the difference between the instruments results from the resolution dependent 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<sub>2</sub> and CH<sub>4</sub> 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 airmasses. 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). However, the effect of local sources/sinks is dampened in the total columns.

Remote sensing has been established as a powerful tool in atmospheric science. Using the sun or moon as a 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 downwards-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<sub>2</sub> and CH<sub>4</sub> with high precision. In order to minimize systematic errors and to achieve the required precision, the column-average dry-air mole fractions (DMF) of CO<sub>2</sub>, named XCO<sub>2</sub> and CH<sub>4</sub>,

named XCH<sub>4</sub>, are determined by normalizing them to the known dry-air mole fractions of O<sub>2</sub> of 20.95 %.

$$X_{\text{CO}_2}(\text{O}_2) = 0.2095 \cdot \frac{\text{column CO}_2}{\text{column O}_2} \quad (1)$$

and

$$X_{\text{CH}_4}(\text{O}_2) = 0.2095 \cdot \frac{\text{column CH}_4}{\text{column O}_2}. \quad (2)$$

All retrieved columns share systematic errors as they are measured by the same observing system. Normalizing them in this way hence reduces some of the systematic errors. As the XCO<sub>2</sub>(O<sub>2</sub>) and XCH<sub>4</sub>(O<sub>2</sub>) is always calculated by ratioing with O<sub>2</sub> in this paper, the (O<sub>2</sub>) is tacitly left out from now on.

These ground-based observations are organized into a global measurement community, the Total Carbon Column Observing Network or 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 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<sup>-1</sup> and 0.02 cm<sup>-1</sup>, respectively, where the resolution is defined (following the Bruker notation) as resolution = 0.9/OPD, where the OPD is the maximum optical path difference. Throughout this paper the standard TCCON retrieval windows for CO<sub>2</sub>, CH<sub>4</sub> and O<sub>2</sub>, as shown in Table 1 have been used.

XCO<sub>2</sub> can be measured with a precision of better than 0.25 % and XCH<sub>4</sub> 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<sub>2</sub> and 0.4 % for XCH<sub>4</sub> (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 measure total columns of CO<sub>2</sub> and CH<sub>4</sub>. 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 traveling 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

**Table 1.** Standard TCCON retrieval windows for CO<sub>2</sub>, CH<sub>4</sub> and O<sub>2</sub>.

Gas	Central wavenumber [cm <sup>-1</sup> ]	Spectral width [cm <sup>-1</sup> ]
CO <sub>2</sub>	6220.00	80.00
CO <sub>2</sub>	6339.50	85.00
CH <sub>4</sub>	5938.00	116.00
CH <sub>4</sub>	6002.00	11.10
CH <sub>4</sub>	6076.00	138.00
O <sub>2</sub>	7885.00	240.00

greenhouse gas instruments SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Cartography) onboard ENVISAT (Environmental Satellite), TANSO-FTS (Thermal And Near Infrared Sensor for Carbon Observation Fourier Transform Spectrometer) onboard GOSAT (Greenhouse Gases Observing Satellite) and OCO-2 (Orbiting Carbon Observatory) are 7.0 cm<sup>-1</sup>, 0.2 cm<sup>-1</sup> and 0.3 cm<sup>-1</sup>, 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<sub>2</sub> and CH<sub>4</sub> using the small commercial interferometer Bruker IFS 66 at its maximum possible resolution of 0.11 cm<sup>-1</sup>. The measurements have been performed in Bremen, Germany (53.1° N, 8.8° E) for nine days between winter 2009 and spring 2010. 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<sup>-1</sup> and 0.5 cm<sup>-1</sup>.

## 2 Instrumentation

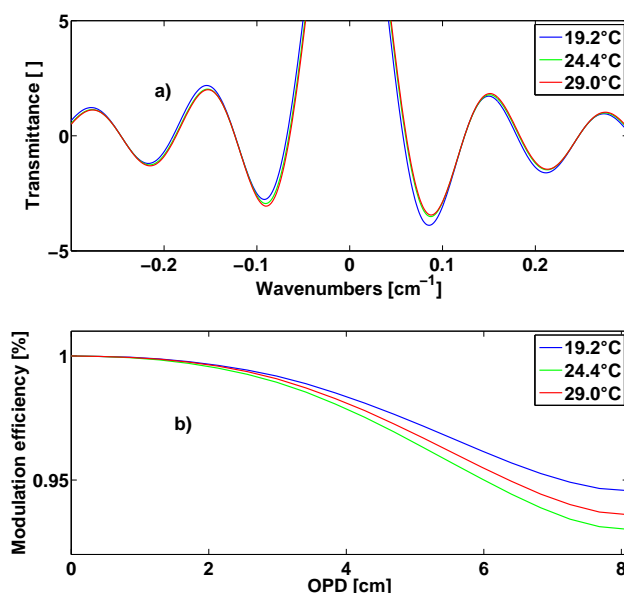
The IFS 66 was installed in a room at our institute above the laboratory where the IFS 125 HR is housed. A home-made solar tracker, driven by a quadrant diode, is installed on the top of the building, feeding a parallel light beam onto the entrance aperture of the IFS 125 HR. In order to minimize the 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. Spectra were recorded with the IFS 66 at a maximum resolution of  $0.11\text{ cm}^{-1}$  and with the IFS 125 HR at  $0.014\text{ cm}^{-1}$ . 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 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<sub>2</sub> beamsplitters and indium gallium arsenide (In-GaAs) 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 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 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.

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 fit the observations by minimizing the RMS residual. The retrieved DMFs of CO<sub>2</sub> and CH<sub>4</sub> are calculated by O<sub>2</sub> as discussed in the introduction. So far it is not possible to consider the detector noise in the GFIT retrieval. In our analysis we used a beta-version of GFIT that has been modified to pre-adjust the stratospheric a-priori concentration of CH<sub>4</sub>, by making use of the known correlation between stratospheric CH<sub>4</sub> 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<sub>4</sub> (Washenfelder et al., 2003). This leads to better spectral fits for CH<sub>4</sub>, as discussed later.

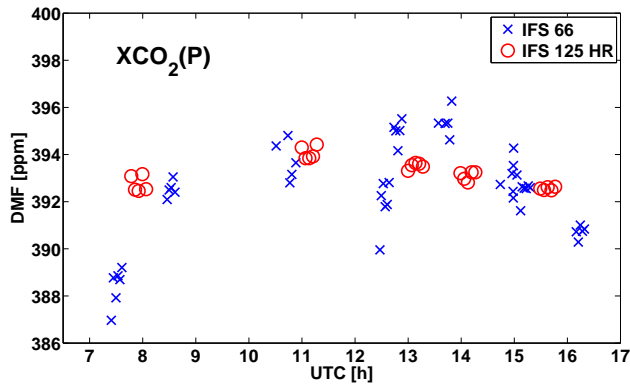


**Fig. 1.** Linefit evaluation with a HCl gas cell for different temperatures. Panel (a) shows the ILS (the line is truncated to show the important part in more detail), (b) shows the modulation efficiency decreasing with the OPD.

## 4 Results

### 4.1 Alignment and stability

The alignment of both instruments is checked by cell measurements using an HCl gas-cell of known pressure and temperature. Hereby, the Instrumental Line Shape (ILS) and the modulation efficiency are retrieved. The modulation efficiency gives the amount of interference of the two light beams within an FTS instrument. The light beam entering the FTS is split at the beamsplitter. The two parts of the beam propagate different ways towards two different mirrors and back, overlapping at the beamsplitter. In a perfectly aligned instrument, the two parts of the beam are perfectly overlapping for the same OPD in both beams (zero path difference), which leads to a fully constructive interference for every wavelength. The modulation efficiency varies as a function of the OPD. In our set up the modulation efficiency is measured by performing cell measurements of a gas with well known pressure and temperature. Then the comparison of the measured and simulated spectrum allows to retrieve the modulation efficiency and the ILS. The instrumental line shape (ILS) is the response we receive with the FTS from a theoretical singular peak at a specific wavenumber. The ILS is a sinc function which should be symmetric. The resolution of the FTIR is mainly determined by the width of the ILS, which is a function of the OPD and the aperture. If a FTS is misaligned, the ILS is lowered, broadened, and the wavenumber is shifted or asymmetric. A parallel offset leads to a lowered ILS, a hereby measured spectrum has a lower signal to noise

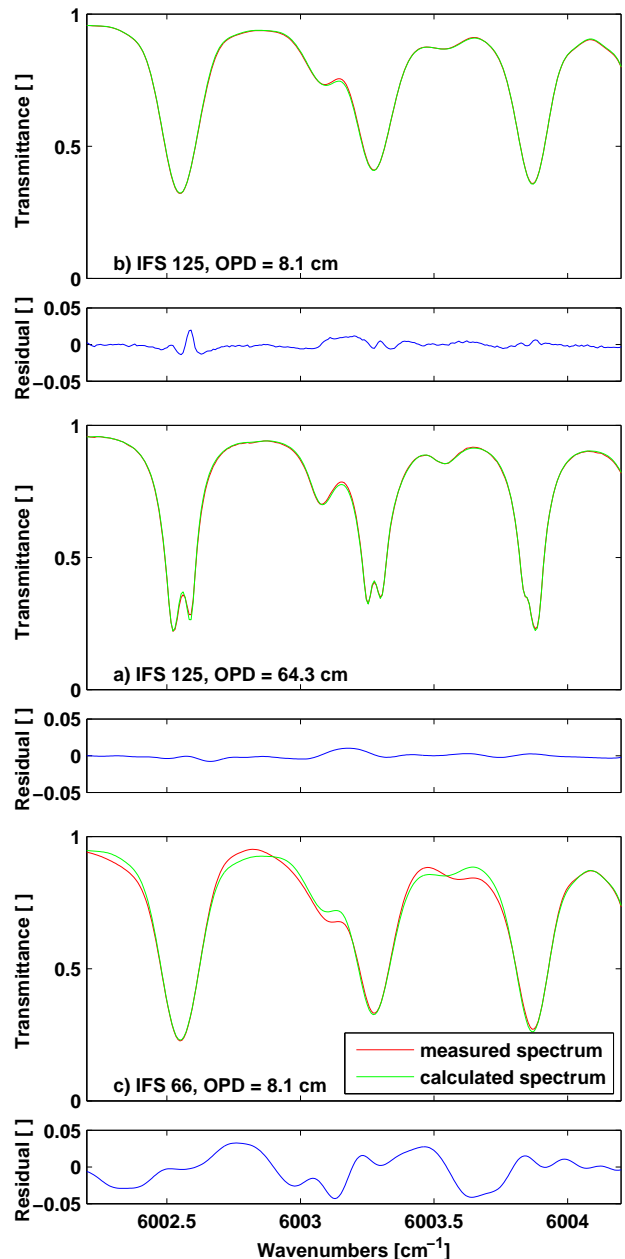


**Fig. 2.** Individual results for column-average dry-air mole fractions (DMF) of CO<sub>2</sub>, as measured by both instruments for 9 March 2010. The shown values are calculated by ratioing to the surface pressure rather than O<sub>2</sub>, as shown in Fig. 4. The SZAs vary from 78 degrees in the morning to 57 degrees at noon and 82 degrees in the afternoon.

ratio. A misalignment at the outgoing part of the FTS leads to a broadened ILS and a lowered resolution. A slope between the overlapping beams leads to an asymmetric ILS and a shift in wavelength. The lowering and the broadening of the ILS is not a major problem as we have enough intensity and high enough resolution. The shift in wavelength could be fitted, mainly the asymmetric ILS could become a serious problem. So far it is not possible to consider the measured ILS in the GFIT retrieval.

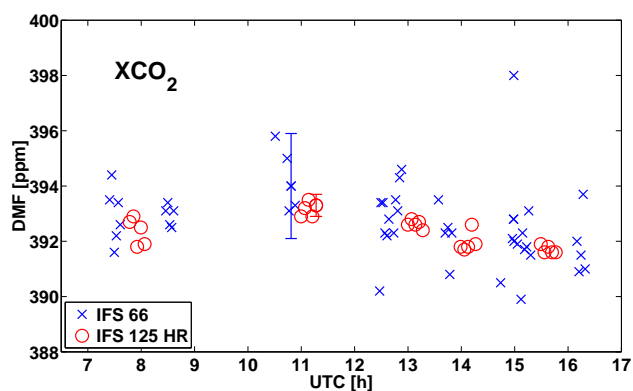
The ILS was retrieved with the “LINEFIT” program using HCl lines from 5680 to 5800 cm<sup>-1</sup> (Hase et al., 1999). The IFS 125 HR typically gives a variation in the modulation efficiency of  $\leq 5\%$  over the full OPD. For the IFS 66, the modulation efficiency that has been measured directly 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 (A. K. Bruker, personal communication, 2009). 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 a change in the modulation efficiency of 2%. We hence performed a test where we took cell measurements at different room temperatures, results are shown in Fig. 1.

The comparison for XCO<sub>2</sub> 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<sub>4</sub> depend on the alignment, see Tables 2 and 3. Tentatively, we assign this dependency to the combination of the least-squares fitting together with

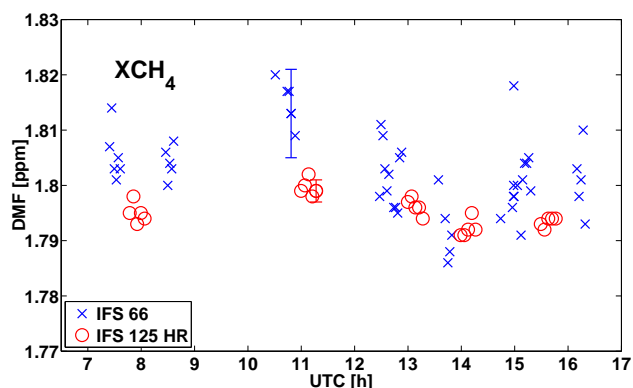


**Fig. 3.** Small part of a measured and simulated spectrum in the region of CH<sub>4</sub>. Panel (a) shows the high resolution spectrum of the IFS 125 HR at a resolution of 0.014 cm<sup>-1</sup> (OPD = 64 cm), (b) is based on the same measurement, but the interferogram has been truncated to a resolution of 0.11 cm<sup>-1</sup> (OPD = 8.1 cm), and (c) shows a spectrum measured by the IFS 66 at a resolution of 0.11 cm<sup>-1</sup>.

an imperfect a-priori VMR. When using a correct a priori, the deviations resulting from a misaligned instrument partly compensate, because the positive and negative residuals have a similar peak height. For an a priori that is far from ideal, the residuals of the fit might have different peak heights. In this



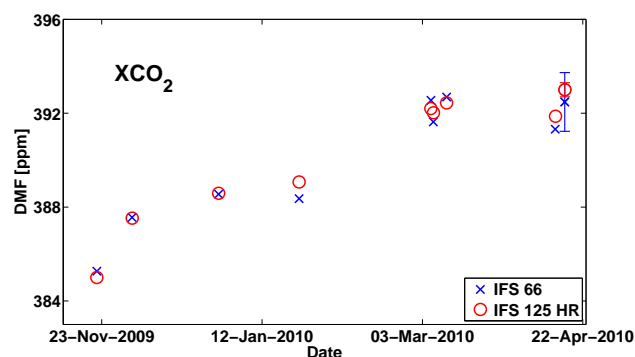
**Fig. 4.** Individual results for column-average dry-air mole fractions (DMF) of CO<sub>2</sub>, as measured by both instruments for 9 March 2010. 10 interferograms have been averaged for the IFS 66 for one data point, and 2 interferograms for the IFS 125 HR. With typical error bars for single measurements as given by the retrieval software GFIT.



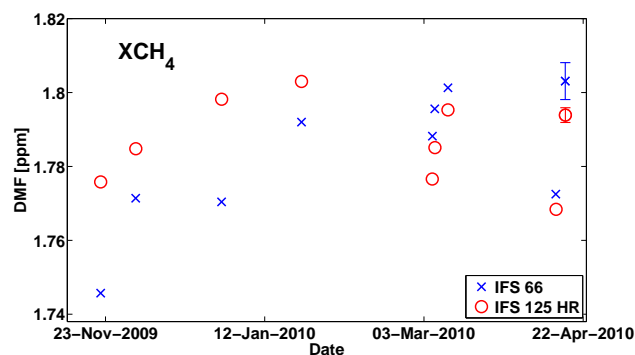
**Fig. 5.** Individual results for column-average dry-air mole fractions (DMF) of CH<sub>4</sub>, as measured by both instruments for 9 March 2010. 10 interferograms have been averaged for the IFS 66 for one data point, and 2 interferograms for the IFS 125 HR. With typical error bars for single measurements as given by the retrieval software GFIT.

case the quadratic dependency of the least-squares method yields wrong columns.

As an example, Fig. 3 shows part of the spectral fit in one CH<sub>4</sub> absorption window. Figure 3a shows the measurements by the IFS 125 HR at a resolution of 0.014 cm<sup>-1</sup> (64.3 cm OPD). Figure 3b shows the same spectrum as in Fig. 3a, but the interferogram has been truncated to 0.11 cm<sup>-1</sup> (8.1 cm OPD), corresponding to the resolution of the IFS 66. Figure 3c shows the spectrum measured by the IFS 66 at a resolution of 0.11 cm<sup>-1</sup> (8.1 cm OPD). The residuals of Fig. 3c have systematic deviations at the absorption lines, which we attribute to non-optimal alignment of the interferometer. Our measurements show that the retrieved values are acceptable for this instrument despite the asymmetric fit as shown in Fig. 3c.



**Fig. 6.** Daily averages of XCO<sub>2</sub>, as measured by the IFS 66 and the IFS 125 HR. The daily means are calculated from single measurements as a weighted mean, weighted to the reciprocal of their error variance. As example typical error bars of one  $\sigma$  are shown.



**Fig. 7.** Daily averages of XCH<sub>4</sub>, as measured by the IFS 66 and the IFS 125 HR. The statistics are calculated as described for XCO<sub>2</sub> in Fig. 6.

## 4.2 Precision and comparability

Results for individual measurements of XCO<sub>2</sub> and XCH<sub>4</sub> are shown in Figs. 4 and 5. The error bars indicate the typical uncertainty for single measurements as given by the retrieval software GFIT. The shown day is selected as it covers a large variation of solar zenith angles (SZA) from 78 degrees in the morning to 57 degrees at noon and 82 degrees in the afternoon. The measurements of XCO<sub>2</sub> and XCH<sub>4</sub> show that there is not a significant dependency on SZAs. For the IFS 66, we obtained a precision of 0.32 % (1.2 ppm) for CO<sub>2</sub> and 0.41 % for CH<sub>4</sub> (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<sub>2</sub> and 0.22 % (3.9 ppb) for CH<sub>4</sub>, 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 2 and 3 give individual results for the nine days on which comparable measurements were possible. In addition to the derived precision the uncertainty based on the spectral fit and residuals is given.

**Table 2.** Daily means of XCO<sub>2</sub> for nine measuring days. The daily means are calculated from single measurements as a weighted mean, weighted to the reciprocal of their error variance. As an estimation for the error of the daily means the standard deviation,  $\sigma$ , is given. The differences of the daily means are given in percent while the averages of differences are calculated from the absolute values.

Day	XCO <sub>2</sub> IFS-125 HR [ppm]	1 $\sigma$ [ppm]	XCO <sub>2</sub> IFS-66 [ppm]	1 $\sigma$ [ppm]	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
Average before alignment	387.55	1.08	387.44	1.21	0.07
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 after alignment	392.31	0.91	392.13	1.25	0.11

**Table 3.** Daily means of XCH<sub>4</sub> for nine measuring days. The statistics are calculated as described for XCO<sub>2</sub> in Table 2. In the last column an offset of 0.47 % is subtracted from the daily means measured by the IFS-66, then the difference is calculated and given in percent. Again, the averages of differences are calculated from the absolute values.

Day	XCH <sub>4</sub> IFS-125 HR [ppm]	$\sigma$ [ $\times 10^{-2}$ ppm]	XCH <sub>4</sub> IFS-66 [ppm]	$\sigma$ [ $\times 10^{-2}$ ppm]	Difference [%]	Diff.-offset [%]
20 November 2009	1.776	0.52	1.746	1.29	-1.69	-2.15
1 December 2009	1.785	0.17	1.771	1.14	-0.78	-1.25
28 December 2009	1.798	0.73	1.770	0.53	-1.56	-2.02
22 January 2010	1.803	0.45	1.792	0.57	-0.61	-1.08
Average before alignment	1.791	0.47	1.770	0.88	1.16	1.62
4 March 2010	1.777	1.11	1.788	0.79	0.62	0.15
5 March 2010	1.785	0.18	1.796	0.79	0.62	0.14
9 March 2010	1.795	0.39	1.801	0.71	0.33	-0.14
12 April 2010	1.768	0.20	1.773	0.83	0.28	-0.19
15 April 2010	1.794	0.07	1.803	0.51	0.50	0.03
Average after alignment	1.784	0.39	1.792	0.73	0.47	0.13

Comparing the daily means from both instruments gives an average deviation of 0.09 % or 0.35 ppm for XCO<sub>2</sub> which is within the precision of both instruments (Fig. 6). Daily means for XCH<sub>4</sub> are shown in Fig. 7. For XCH<sub>4</sub> the results from the IFS 66 are lower by 0.47 % 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 0.13 %, in agreement with the precision of both instruments.

Till the 22 January 2010, the IFS 66 was not well aligned, so the effect of an improperly aligned instrument on the resulting XCO<sub>2</sub> and XCH<sub>4</sub> could be tested by comparing the results.

While XCO<sub>2</sub> is only slightly affected by a bad alignment, the deviations for XCH<sub>4</sub> increase by a factor of three, as given in Tables 2 and 3.

Evaluating the total columns of CO<sub>2</sub> and CH<sub>4</sub> shows similar results. O<sub>2</sub> and CO<sub>2</sub> agree within the error bars, expect for very high SZAs in the early morning and late evening. In the ratio this deviation is canceled out. Figure 2 shows the retrieved XCO<sub>2</sub> calculated by ratioing to the surface pressure column rather than measured O<sub>2</sub> as is shown in Fig. 4, showing the effect of the ratioing to O<sub>2</sub>. CH<sub>4</sub> differs in the same way as XCH<sub>4</sub> with additional errors for very high SZAs. The difference of CO<sub>2</sub> and CH<sub>4</sub> as measured by both instruments do not depend on the SZA.

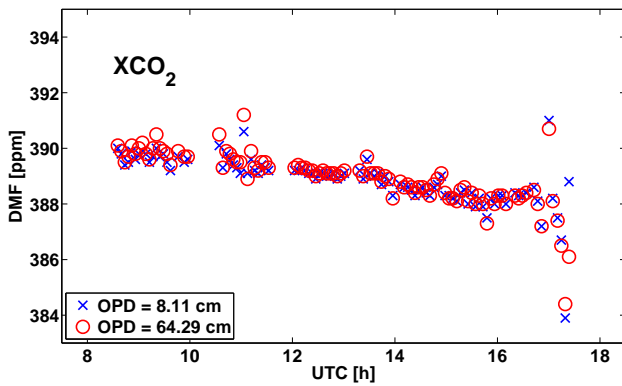


Fig. 8. Retrieved XCO<sub>2</sub> from observations by the high resolution instrument IFS 125 HR for the 16 April 2009. The red symbols correspond to the high resolution of 0.014 cm<sup>-1</sup>, the blue symbols to a resolution of 0.11 cm<sup>-1</sup>.

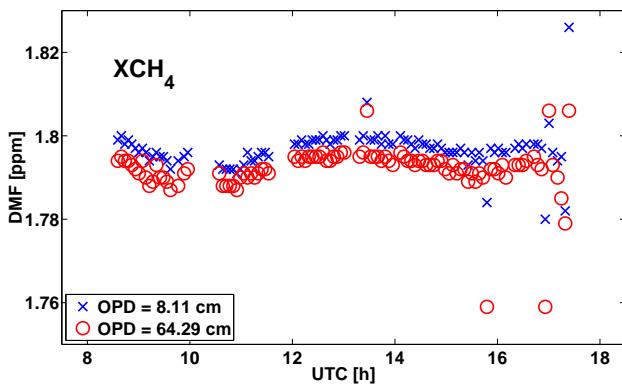


Fig. 9. Retrieved XCH<sub>4</sub> from observations by the high resolution instrument IFS 125 HR for the 16 April 2009. The red symbols correspond to the high resolution of 0.014 cm<sup>-1</sup>, the blue symbols to a resolution of 0.11 cm<sup>-1</sup>.

### 4.3 Influence of resolution

When comparing XCH<sub>4</sub> obtained from both instruments, we attribute the offset of 0.47 % to a combination of improper a-priori profiles, erroneous spectroscopic data, and/or incorrect assumptions in the pT-profile in the retrieval. The residuals in Fig. 3 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 0.11 cm<sup>-1</sup>, the resolution used by the IFS 66. The results are shown in Figs. 8 and 9. While XCO<sub>2</sub> shows almost no dependence on the resolution (0.02 %), for XCH<sub>4</sub> a constant offset of 0.26 % can be observed. The remaining 0.21 % are within the uncertainties for XCH<sub>4</sub> and we get agreement with our estimation given above.

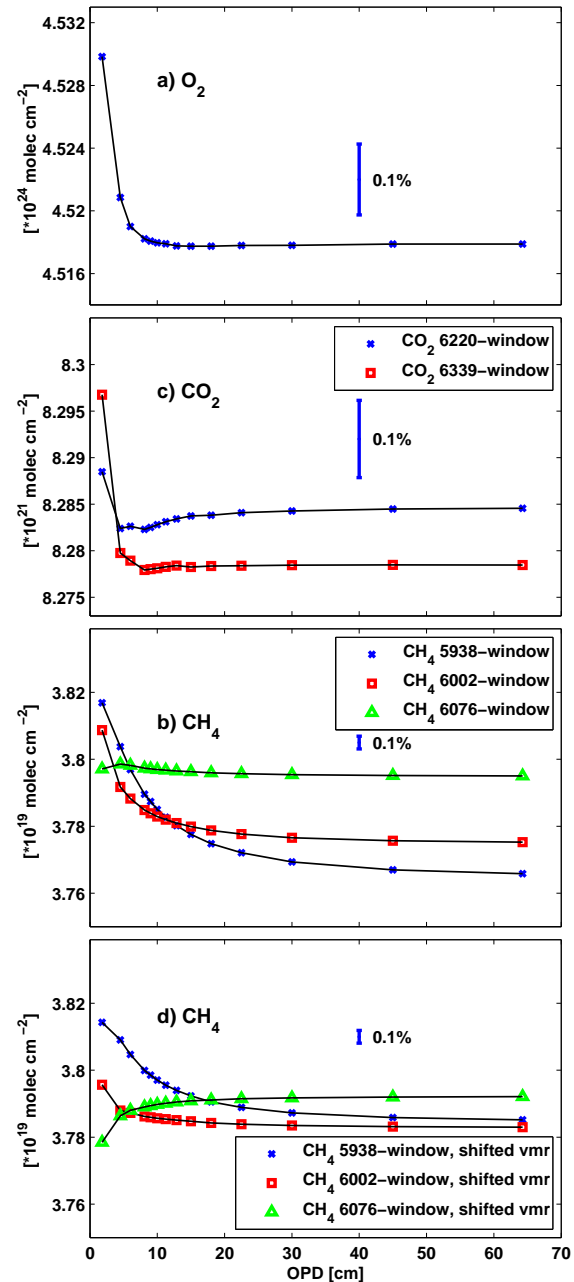
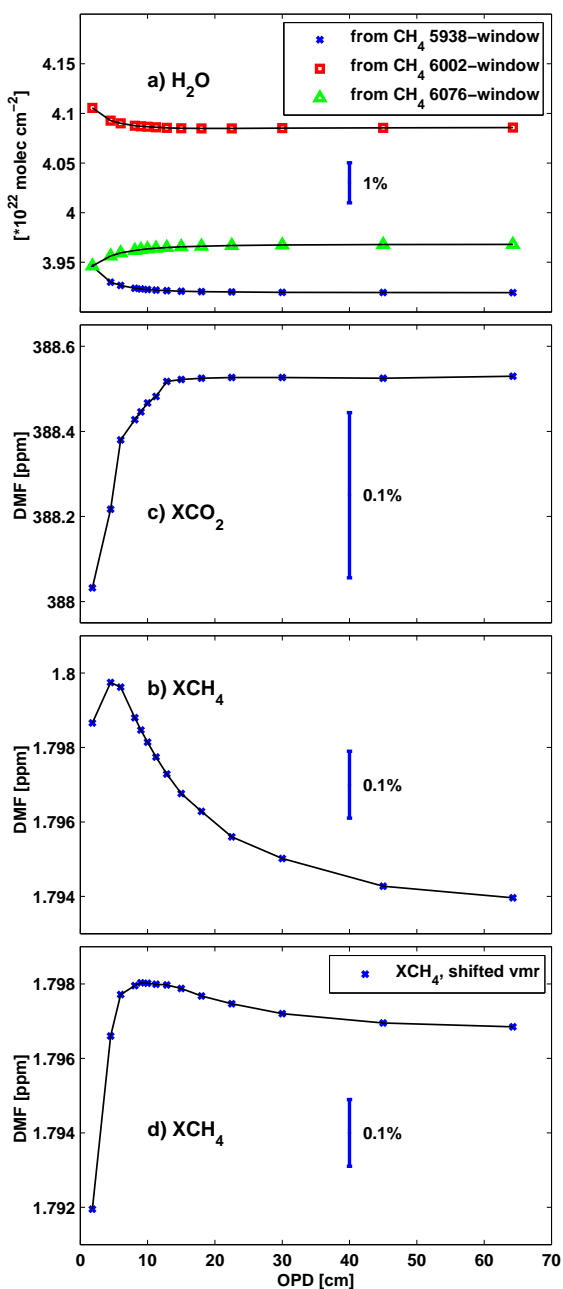


Fig. 10. Retrieved O<sub>2</sub>, CO<sub>2</sub> and CH<sub>4</sub> total columns from the IFS 125 HR, by truncating the interferograms to different optical path differences. For CO<sub>2</sub> and CH<sub>4</sub> the different retrieval windows are shown. For CH<sub>4</sub> the initial a-priori profile has been shifted vertically by -4 km. The shown bars give a percentage scale.

In order to investigate this in more detail, we truncated the high resolution interferograms of the IFS 125 HR in several steps from a resolution of 0.014 cm<sup>-1</sup> (64.3 cm OPD) down to a resolution of 0.5 cm<sup>-1</sup> (1.8 cm OPD), which covers the range of the resolution of the satellite spectrometers OCO-2 and TANSO-FTS/GOSAT. The results are shown in Figs. 10a-c and 11a-c. The tests have been performed for all spectra



**Fig. 11.** Retrieved H<sub>2</sub>O, XCO<sub>2</sub> and XCH<sub>4</sub> from the IFS 125 HR, H<sub>2</sub>O is taken from the different retrieval windows of CH<sub>4</sub>. For XCH<sub>4</sub> the initial a-priori profile has been shifted vertically by  $-4$  km. The shown bars give a percentage scale.

measured on 16 April 2009. Shown is the daily mean. For reference, the variability in percent for each compound is given as a bar in the figure. The dependence on the OPD increases with decreasing OPD. Down to an OPD of 6 cm (resolution of  $0.15\text{ cm}^{-1}$ ), the O<sub>2</sub> and CO<sub>2</sub> columns show only a weak dependency of 0.02 %. For CH<sub>4</sub> columns, the variation is much larger, between 0.05 % and 0.3 %, depending on the spectral band analyzed. The corresponding results for

XCO<sub>2</sub>, XCH<sub>4</sub> and the most important interfering gas, H<sub>2</sub>O, are also shown. The dependency for H<sub>2</sub>O is about 0.1 %. As there are H<sub>2</sub>O lines in the window where CH<sub>4</sub> is retrieved ( $5938\text{ cm}^{-1}$ ,  $6002\text{ cm}^{-1}$  and  $6076\text{ cm}^{-1}$ ), for low resolution spectra the lines cannot be separated.

These simulations for XCO<sub>2</sub> and XCH<sub>4</sub> are in agreement with the results from the comparison of both instruments, as shown in Figs. 4 and 5. While the CO<sub>2</sub> total columns for both instruments agree within the precision, for CH<sub>4</sub>, a constant bias of 0.26 % was found. It has to be mentioned that standard TCCON measurements are made with an OPD of 40 cm and are not significantly affected.

In order to investigate the dependency of CH<sub>4</sub> on the OPD in more detail, we shifted the whole a-priori VMR profile of CH<sub>4</sub> down by 4 km, and repeated the resolution-dependant sensitivity study shown in Figs. 10c and 11c. The results are displayed in Figs. 10d and 11d. The simulations show that the dependency of the resolution on the CH<sub>4</sub> total column is less. Furthermore, the dependency of the windows at  $6002\text{ cm}^{-1}$  and  $6076\text{ cm}^{-1}$  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, 4 km yielded the smallest dependency on the OPD. A downshifting of the a-priori VMR by 4 km is certainly not realistic, but our results might indicate that the a priori and/or the spectral data of CH<sub>4</sub> are erroneous. For completeness, we have repeated the simulations for CO<sub>2</sub> and O<sub>2</sub> 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.

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<sub>4</sub> 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 ( $0.15\text{ cm}^{-1}$ ) 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.



## 5 Summary and conclusion

The precision for the low resolution instrument was found to be 0.32 % for XCO<sub>2</sub> (1.2 ppm) and 0.41 % (7.3 ppb) for XCH<sub>4</sub>. For comparable measurement times, the precision of the IFS 66 is lower compared to the IFS 125 HR, which can be explained 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<sub>2</sub>. For XCH<sub>4</sub> a constant offset of 0.47 % has been observed. After subtracting the constant offset, the average deviation between the instruments was also of the order of 0.2 % (4 ppb). 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<sub>4</sub>. The ILS of the low resolution instrument was found to depend on the temperature. While the results for XCO<sub>2</sub> do not depend on the temperature and alignment, for XCH<sub>4</sub> the precision and comparability require a well aligned instrument. Overall we can conclude that XCO<sub>2</sub> can be measured with a low resolution FTS instrument with sufficient precision and accuracy, however, for XCH<sub>4</sub> the results depend on the alignment.

*Acknowledgements.* We acknowledge the support by the European Commission of the Integrated Project InGOS (Integrated non-CO<sub>2</sub> Greenhouse gas Observation System) within the 7th Framework Program. We acknowledge the ESA GHG CCI (European Space Agency Green House Gas Climate Change Initiative) project. We thank Nick Deutscher for his comments on the manuscript. This work was financially supported by the University of Bremen and the Senate of Bremen.

Edited by: I. Aben

## References

- Deutscher, N. M., Griffith, D. W. T., Bryant, G. W., Wennberg, P. O., Toon, G. C., Washenfelder, R. A., Keppel-Aleks, G., Wunch, D., Yavin, Y., Allen, N. T., Blavier, J.-F., Jiménez, R., Daube, B. C., Bright, A. V., Matross, D. M., Wofsy, S. C., and Park, S.: Total column CO<sub>2</sub> measurements at Darwin, Australia – site description and calibration against in situ aircraft profiles, *Atmos. Meas. Tech.*, 3, 947–958, doi:10.5194/amt-3-947-2010, 2010.
- Hase, F., Blumenstock, T., and Paton-Walsh, C.: Analysis of the instrumental line shape of high-resolution Fourier transform IR spectrometers with gas cell measurements and new retrieval software, *Appl. Optics*, 38, 3417–3422, doi:10.1364/AO.38.003417, 1999.
- Keppel-Aleks, G., Toon, G. C., Wennberg, P. O., and Deutscher, N. M.: Reducing the impact of source brightness fluctuations on spectra obtained by Fourier-transform spectrometry, *Appl. Optics*, 46, 4774–4779, 2007.
- Messerschmidt, J., Macatangay, R., Notholt, J., Petri, C., Warneke, T., and Weinzierl, C.: Side by side measurements of CO<sub>2</sub> by ground-based Fourier transform spectrometry (FTS), *Tellus B*, 62, 749–758, 2010.
- Messerschmidt, J., Geibel, M. C., Blumenstock, T., Chen, H., Deutscher, N. M., Engel, A., Feist, D. G., Gerbig, C., Gisi, M., Hase, F., Katrynski, K., Kolle, O., Lavrič, J. V., Notholt, J., Palm, M., Ramonet, M., Rettinger, M., Schmidt, M., Sussmann, R., Toon, G. C., Truong, F., Warneke, T., Wennberg, P. O., Wunch, D., and Xueref-Remy, I.: Calibration of TCCON column-averaged CO<sub>2</sub>: the first aircraft campaign over European TCCON sites, *Atmos. Chem. Phys.*, 11, 10765–10777, doi:10.5194/acp-11-10765-2011, 2011.
- Petersen, A. K., Warneke, T., Frankenber, C., Bergamaschi, P., Gerbig, C., Notholt, J., Buchwitz, M., Schneising, O., and Schrems, O.: First ground-based FTIR observations of methane in the inner tropics over several years, *Atmos. Chem. Phys.*, 10, 7231–7239, doi:10.5194/acp-10-7231-2010, 2010.
- Toon, G. C., Farmer, C. B., Schaper, P. W., Lowes, L. L., and Norton, R. H.: Composition measurements of the 1989 arctic winter stratosphere by airborne infrared solar absorption spectroscopy, *J. Geophys. Res.*, 97, 7939–7961, doi:10.1029/91JD03114, 1992.
- Warneke, T., Yang, Z., Olsen, S., Korner, S., Notholt, J., Toon, G. C., Velasco, V., Schulz, A., and Schrems, O.: Seasonal and latitudinal variations of column averaged volume-mixing ratios of atmospheric CO<sub>2</sub>, *Geophys. Res. Lett.*, 32, L03808, doi:10.1029/2004GL021597, 2005.
- Washenfelder, R. A., Wennberg, P. O., and Toon, G. C.: Tropospheric methane retrieved from ground-based near-IR solar absorption spectra, *J. Geophys. Res.*, 30, 2226, doi:10.1029/2003GL017969, 2003.
- Washenfelder, R., Toon, G., Blavier, J., Yang, Z., Allen, N., Wennberg, P., Vay, S., Matross, D., and Daube, B.: Carbon dioxide column abundances at the Wisconsin Tall Tower site, *J. Geophys. Res.*, 111, D22305, doi:10.1029/2006JD007154, 2006.
- Wunch, D., Toon, G. C., Wennberg, P. O., Wofsy, S. C., Stephens, B. B., Fischer, M. L., Uchino, O., Abshire, J. B., Bernath, P., Biraud, S. C., Blavier, J.-F. L., Boone, C., Bowman, K. P., Browell, E. V., Campos, T., Connor, B. J., Daube, B. C., Deutscher, N. M., Diao, M., Elkins, J. W., Gerbig, C., Gottlieb, E., Griffith, D. W. T., Hurst, D. F., Jiménez, R., Keppel-Aleks, G., Kort, E. A., Macatangay, R., Machida, T., Matsueda, H., Moore, F., Morino, I., Park, S., Robinson, J., Roehl, C. M., Sawa, Y., Sherlock, V., Sweeney, C., Tanaka, T., and Zondlo, M. A.: Calibration of the Total Carbon Column Observing Network using aircraft profile data, *Atmos. Meas. Tech.*, 3, 1351–1362, doi:10.5194/amt-3-1351-2010, 2010.
- Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B., Griffith, D., and Wennberg, P. O.: The Total Carbon Column Observing Network (TCCON), *Philos. T. R. Soc. A.*, 369, 1943, doi:10.1098/rsta.2010.0240, 2011.
- Yang, Z. H., Toon, G. C., Margolis, J. S., and Wennberg, P. O.: Atmospheric CO<sub>2</sub> retrieved from ground-based near IR solar spectra, *Geophys. Res. Lett.*, 29, 1339, doi:10.1029/2001GL014537, 2002.
- Yang, Z. H., Washenfelder, R. A., Keppel-Aleks, G., Krakauer, N. Y., Randerson, J. T., Tans, P. P., Sweeney, C., and Wennberg, P. O.: New constraints on northern hemisphere growing season net flux, *Geophys. Res. Lett.*, 34, L12807, doi:10.1029/2007GL029742, 2007.