

FTS measurements of column CO₂ at Sodankylä

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Abstract

Fourier Transform Spectrometer (FTS) observations at Sodankylä, Finland (67.4° N, 26.6 ° E) have been performed since early 2009. The FTS instrument is participating in the Total Carbon Column Observing Network (TCCON) and has been optimized to measure abundances of the key greenhouse gases in the atmosphere. Sodankylä is the only TCCON station in the Fennoscandia region. Here we report the measured CO₂ time series over a seven year period (2009-2015) and provide a description of the FTS system and data processing at Sodankylä. We find the lowest monthly column CO₂ values in August and the highest monthly values during the February to May season. Inter-annual variability is the highest in June-September period, which correlates with the growing season. During the time period of FTS measurements from 2009 until 2015 we have observed a 2.2+/-0.2 ppm increase per year in column CO₂. The monthly mean column CO₂ values have exceeded 400 ppm level for the first time in February 2014.

1 Introduction

Carbon dioxide (CO₂) is the most abundant anthropogenic greenhouse gas in the atmosphere (Hartman et al., 2013). The concentration of CO₂ has increased due to the burning of carbon-based fuels. Precise and accurate measurements of CO₂ are needed in order to better understand the carbon cycle. In addition to the relatively long term *in situ* measurements of CO₂, ground based total column measurements of carbon dioxide have become possible more recently. The column averaged dry-air mole fractions of carbon dioxide (XCO₂) have been measured since year 2004 by the Total Carbon Column Observing Network (TCCON) sites, using solar Fourier Transform Spectrometers (FTS), operating in the near infrared spectral

region (Wunch et al., 2011a). The main goal of the TCCON has been to provide precise and accurate measurements of XCO₂, but also other gases have been retrieved, including CH₄, CO, N₂O, H₂O, HDO and HF. Compared to the surface *in situ* measurements XCO₂ is less affected by changes in the height of planetary boundary layer and the spatial sensitivity footprint is larger (Keppel-Aleks et al., 2011). The accuracy and precision of the XCO₂ measurements within TCCON is better than 0.25% (Wunch et al., 2011a). The high accuracy and precision is needed to contribute to the carbon cycle research and validation of space borne measurements. Satellite missions that have already used the TCCON data include the Orbiting Carbon Observatory-2 (OCO-2; Crisp et al., 2004); the Greenhouse Gases Observing Satellite (GOSAT; Yokota et al., 2009) and the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY; Bovensmann et al., 1999).

Sodankylä in northern Finland is one of the stations in the TCCON. This is currently the only TCCON station in the Fennoscandia region. We established the FTS measurements at Sodankylä in early 2009. Since then the XCO₂ retrievals have been used in several studies (e.g. in Wunch et al., 2011b; Oshchepkov et al., 2012; Saito et al., 2012; Belikov et al., 2013; Guerlet et al., 2013; Yoshida et al., 2013; Agustí-Panareda, 2014; Deng et al., 2014; Reuter et al., 2014; Barthlott et al., 2015; Heymann et al., 2015; Lindqvist et al., 2015; Belikov et al., 2016; Feng et al., 2016; Inoue et al., 2016; Massart et al., 2016). This paper describes the instrumentation, measurement procedures and data processing at the Sodankylä FTS site, corresponding to the data retrieval version GGG2014 (Wunch et al., 2015). The quality controlled data from May 2009 until November 2015 have been used here to calculate the average seasonal cycle and trend of XCO₂ over the measurement period.

2 Instrumentation

The Sodankylä TCCON FTS station is part of the infrastructure of the Finnish Meteorological Institute's Arctic Research Center. The FTS is located at 67.3668° N, 26.6310° E, 188 m.a.s. FTS measurements at Sodankylä are made using a Bruker 125 HR FTS (Bruker Optics, Germany). Since the beginning of the data record the FTS instrument has been installed in a two-story observational building. The interior of the laboratory was rebuilt in late 2008 to mount the FTS instrument. The instrument is placed on a concrete plate, which is designed to absorb possible vibration. The solar tracker on the roof of the building is of type A547N,

1 manufactured by Bruker Optics. The cover of the tracker was built locally at the institute's
2 workshop.

3 The FTS instrument is equipped with two room temperature detectors: an indium gallium
4 arsenide (InGaAs, covers 4000-11000 cm^{-1}) and a silicon diode (Si, covers 9000-15000 cm^{-1}),
5 which is similar to the other FTS stations in the TCCON network. The measurements are
6 performed in vacuum to improve stability and to reduce water vapor in the system. The
7 system is evacuated each night to avoid vibration during the solar measurements. The optical
8 path difference (OPD) is 45 cm and the spectral resolution is 0.02 cm^{-1} , collection time for a
9 single scan is 78 seconds.. Column abundances of CO_2 , O_2 , CH_4 , H_2O , HDO , HF , CO and
10 N_2O are retrieved from the spectra.

11 The FTS instrument has worked in a fully automated mode since July 2013. Readings from
12 rain and direct solar radiation sensors, combined with the automated analysis of weather radar
13 forecast data, determine the start and cessation of daily measurements. A control system
14 monitors the measurement quality and automatically reports on error conditions, thus longer
15 measurement gaps have been minimized. Currently used settings are presented in Table 1. In
16 addition to the TCCON measurements, we also take longer wavelength measurements, using
17 a liquid nitrogen cooled indium antimonide detector (InSb, covers 1800 –6000 cm^{-1}). The InSb
18 measurements are filtered, the pass-band is at 2439-3125 cm^{-1} . This filter choice is designed
19 for profile retrievals of methane and provides a possibility to compare the mid infrared (MIR)
20 and near infrared (NIR) retrievals of CH_4 . The sequence of measurements is such that after
21 two InGaAs/Si scans, one InSb scan is taken. To be able to make the solar intensity variation
22 correction, we have recorded all interferograms in the DC mode.

23 To guarantee the optimal performance of the instrument, the optical alignment is checked and
24 adjusted at least once per year. Usually the alignment is performed in winter, because then the
25 solar measurements are not possible due to the high latitude location of the station. We have
26 applied the alignment procedure developed by Hase and Blumenstock (2001). The alignment
27 method is based on the inspection of laser fringes through a telescope. In addition we monitor
28 the instrument line shape (ILS) by taking HCl reference gas measurements on a monthly
29 basis. The ILS retrievals are made using the LINEFIT14 software (Hase et al., 2013). Figure 1
30 presents a selection of ILS retrievals. The upper panel corresponds to the amplitude of the
31 modulation and the lower panel to the phase error, both as functions of optical path
32 difference. Modulation amplitude for a well-aligned FTS should be in the limits of 5% loss at

maximum optical path difference (Wunch et al., 2011a). In Sodankylä case the spread of the values of modulation amplitude is within 3 %, which is very close to the ideal value. The phase error values are measured as being close to zero (Figure 1, lower panel). A small increase in phase error was an indication of temporary scanner problems in July 2012. In general the temporal variability of the modulation efficiency is caused by the scanner wear and slight mechanical influences, which are related to small variabilities in temperature and pressure. This level of small disturbances from the ideal value of modulation efficiency is common to all well aligned spectrometers (Hase et al., 2013). Figure 1 shows that the derived modulation efficiency at maximum OPD has remained relatively stable over time, indicating that the alignment has been maintained.

3 Data processing and availability

Using the InGaAs detector, XCO₂ values are retrieved in two bands, centered at 6228 cm⁻¹ and 6348 cm⁻¹. Within TCCON, the retrieval of XCO₂ and other gases is based on the GFIT algorithm as described by Wunch et al. (2011a). The data processing and analysis scheme is common at each TCCON site, although some sites may have slightly different setup of instrumentation. For example, not all the TCCON stations have the Si detector available.

XCO₂, the column-averaged dry-air mole fraction of CO₂, is defined as the ratio of CO₂ total column to the total column of all gases, excluding water. The total dry air column can be calculated either from surface pressure and water vapor column or from oxygen column, assuming the constant dry-air mole fraction of 20.95% for O₂. The oxygen column is retrieved from the TCCON FTS spectra and the method via oxygen is adopted in TCCON. XCO₂ is the ratio of CO₂ column to O₂ column,

$$XCO_2 = \frac{CO_2 \text{ column}}{O_2 \text{ column}} \times 0.2095 \quad (1)$$

By calculating the ratio, all errors that affect both columns in the same way cancel. This improves the repeatability of the XCO₂ retrieval.

The multiyear data have been reprocessed using the most recent analysis software GGG2014 (Wunch et al., 2015). From the point of view of the historical data homogenization, one of the major improvements in GGG2014 from GGG2012 is the laser sampling error (LSE) correction, which makes use of the simultaneously measured Si spectra. The LSE correction derives the laser sampling errors from Si detector measurements and resamples the

interferograms. In our data record such corrections have been necessary for measurements taken prior to March 3, 2010. Figure 2 shows the time series of the LSE derived from the Si spectra at Sodankylä. In an ideal case the LSE is small and centered around zero. Errors in the sampling of the metrology laser have been caused by faulty electronic boards in the Bruker FTS. These boards were replaced twice in case of our instrument. The ECL02 board was installed on March 10, 2010, and was replaced a year later (Table 2). The currently used electronic board (ECL05) has been operational since March 3, 2011. Intermittent fluctuations in LSE from August 27 until November 11, 2012 and again from July 6 until August 1, 2013 can be explained by scanner problems. The displacement sensor on the scanner positioning board caused fluctuations in scanner moving speed. The positioning board was replaced August 2, 2013 and since then the sampling errors have been minimal.

Another important measure of data quality and instrument performance is xAIR, the column average dry air mole fraction of dry air (Wunch et al., 2015). xAIR is the ratio of total dry air column, calculated from the surface pressure (P_s) and the measured XH_2O , to the total dry air column, obtained from the measured oxygen column:

$$xAIR = \frac{AIR\ column}{O_2\ column} \times 0.2095 - XH_2O \times \frac{m_{H_2O}^{dry}}{m_{air}^{dry}} \quad (2)$$

$$AIR\ column = \frac{P_s}{\{g\}_{air} \times \frac{m_{air}^{dry}}{N_A}} \quad (3)$$

m_{H_2O} and m_{air}^{dry} are the molecular masses of water vapor and dry air, N_A is Avogadro's constant and $\{g\}_{air}$ is the column-averaged gravitational acceleration. Ideally this ratio should be 1, but typically the xAIR value is little less, around 0.98, in TCCON measurements, related to errors in the O_2 spectroscopy (Washenfeller et al., 2006). In practice xAIR is a measure of how well the instrument is capable of obtaining the oxygen column. Large differences in xAIR values compared to the network wide mean are a sign of instrument problems. The problems may be related to several factors, such as a poor optical alignment, spectral ghosts or faulty pressure sensor.

The time series of xAIR are shown in Figure 3. The average xAIR value for 2009-2011 is 0.980 and the average xAIR for the time period of 2012-2015 is 0.978. The first 3 years, until 2012, correspond to the original alignment by Bruker, while the realignment since 2012 was performed using the fringe method. The method is considered an improvement over the original alignment (Hase and Blumenstock, 2001; Heikkinen et al., 2012).

1 The xAIR record shows that the instrument has been stable during its history. xAIR behaves
2 consistently also during the period of relatively large sampling errors, because of the
3 resampling included in the GGG2014 processing scheme. This was not the case with the
4 previous version of data reprocessing system, GGG2012. In the previous data version the
5 xAIR level was too low for the given period of measurements. During the first months of year
6 2009 we didn't have a dichroic beamsplitter installed and therefore we had no Si
7 measurements. Reprocessing the earliest data, from the time period 6.2.2009-15.5.2009 needs
8 a different approach (Dohe et al., 2013). Therefore the data from this time period have not
9 been reprocessed using GGG2014. For the previous data version (GGG2012) we have made
10 an additive LSE correction for the given time period, based on the data collected at different
11 scanner speeds. Without any LSE correction the xGAS values are too low for these months by
12 amounts ranging from 0.2 to 1.0 %. The calculated additive correction for XCO₂ is 2.5 ppm.
13 For other gases the correction is as follows: XCO 0.86 ppb, XCH₄ 0.012 ppm, XH₂O 2.9 ppm
14 and XN₂O 2.4 ppb.

15 The GGG2014 data version in this study covers the time period of 15.5.2009 until 5.11.2015.
16 During these years we have collected 111825 individual measurements, which have been
17 spread over 966 days (Figure 4). If we add the GGG2012 data version from 6.2.2009 until
18 16.5.2009, then the total number of measurement is 123715 (1022 measurement days). A
19 single measurement was graded as acceptable if the solar intensity variation during the
20 measurement was less than 5% and the solar zenith angle was less than 82 degrees. Due to the
21 zenith angle constraint good measurements are only possible from February 8 to November
22 11 each year (268 days) resulting in a gap in winter that is over 3 months long. On average
23 there have been 146 measurement days per year. The main factor that limits the amount of
24 measurements is cloudiness though measurement gaps also occur due to technical problems.
25 A one month gap in the measurements was caused by the failure of sampling laser on May 20,
26 2012; the laser was replaced on June 20, 2012. A slight increase in the amount of
27 measurements can be observed in 2013, because this was the first year when the instrument
28 worked in fully automatic mode.

29 The reprocessed GGG2014 data version of the Sodankylä FTS measurements is available
30 from the Carbon Dioxide Information Analysis Center (Kivi et al., 2014).

31

4 XCO₂ time series and the annual cycle

The absolute values of each of the XCO₂ measurements are presented in Figure 5 (upper panel), corresponding to the time period of 2009-2015. We have also included time series of other gases that are retrieved together with the XCO₂, using GGG2014. The other time series are for XCH₄, XN₂O, XCO, XH₂O and XHF measurements. The non-CO₂ TCCON measurements from Sodankylä have been previously published by e.g. Saito et al. (2012); Belikov et al. (2013); Yoshida et al. (2013); Saad et al. (2014); Tsuruta et al. (2015); Inoue et al. (2016).

Over the seven year time period the trend of XCO₂ is found to be 2.2 \pm 0.2 ppm/year (\pm one standard error). In Figure 6 monthly mean values are plotted for each month when measurements have been possible. The trend is in broad agreement with earlier studies (e.g. Lindqvist et al., 2015), though it is based on a longer time period. It is noteworthy that in February 2014 the monthly mean XCO₂ values have 400 ppm level for the first time, while individual measurements have achieved the 400 ppm level already in spring 2012 and 2013. Similar to the XCO₂, we find significant trend in XCH₄. In case of XCH₄ the observed increase has been 7.1 \pm 0.8 ppb/year.

The average annual cycle of XCO₂ is shown in Figure 7, based on 7 years of measurement. The highest values of XCO₂ are obtained in February to May period, before the start of the growing season. The minimum monthly XCO₂ occurs in August due to the uptake of carbon into the biosphere, which correlates with the period of plant growth. The interannual variability is found to be the smallest in spring (March-May) and largest in summer and autumn (June to September). The shape of the annual cycle can be explained by the imbalance between ecosystem respiration and gross primary production. This is often referred to as net ecosystem exchange (NEE). At high latitudes a negative NEE is observed during the growing season, because the gross primary production has a peak around the summer solstice, while ecosystem respiration has a maximum later in summer, in correlation with the increase in ground and air temperature (Lloyd and Taylor, 1994). Based on the TCCON measurements, Wunch et al. (2013) found that the minima in XCO₂ annual cycle is correlated with summertime surface temperature anomalies. The amplitude of the column CO₂ seasonal cycle at high latitudes of the Northern Hemisphere is smaller than the one based on surface measurement (Olsen and Randerson, 2004). Column CO₂ seasonal variability can be explained by the variability in the terrestrial biospheric fluxes (Keppel-Aleks et al., 2011),

while the long-term trend is driven by the fossil fuel emissions (Hartman et al., 2013). CarbonTracker (Peters et al., 2007) has been widely used to study the annual cycle of XCO₂. It has been shown that CarbonTracker is able to simulate the seasonal cycle at Sodankylä with an average model-measurement bias less than 0.4 ppm (Reuter et al., 2014). Recently the daily forecasts of CO₂ have also become available through Monitoring of Atmospheric Composition and Climate - Interim Implementation (MACC-II) service at the European Centre for Medium- Range Weather Forecasts. The model includes also the short term meteorological variability (Agustí-Panareda et al., 2014).

5 Conclusions and outlook

XCO₂ measurements have been made at Sodankylä since early 2009. The FTS instrument has been relatively stable. Regular instrument alignments and HCl cell measurements have been performed. The instrument has run in fully automatic mode since 2013, therefore the temporal data coverage is relatively good, given the high latitude conditions at Sodankylä. The historical data have been reprocessed using the GGG2014 software (Wunch et al., 2015). The data have been made available via the Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA (Kivi et al., 2014). Measurements from other TCCON sites are also available from the same data center.

Based on the measurements at Sodankylä we find a 2.2±0.2 ppm increase per year in XCO₂ values. In February 2014 the monthly mean XCO₂ values have exceeded 400 ppm level for the first time in the history of these measurements. The lowest monthly XCO₂ values within the seasonal cycle are found in August and the highest in February-May. Year-to-year variability is lowest in March-May and highest during the growing season in June-September.

Relevant to the FTS measurements, we have started with balloon borne AirCore (Karion et al., 2010) profile measurements of CO₂, CH₄ and CO at Sodankylä in September 2013. The balloon measurements have the benefit of reaching much higher vertical altitudes (up to 30-35 km), compared to the aircraft *in situ* measurements. In addition, year around measurements by AirCore are possible. AirCore used in Sodankylä is a 100 m long coiled sampling tube, with a volume of ≈ 1400 ml (Paul et al., 2016). The sampling tube is filled during the payload descent and is automatically closed 9 seconds after the landing. The profile analysis has been performed typically within 2-3 hours after the landing of the payload. Gas analysis have been performed by a Cavity Ring-Down Spectrometer (Picarro Inc., CA, model G2401). Total gas

column measured by an AirCore sampling system is directly related to the World Meteorological Organization *in situ* trace gas measurement scales. Therefore the measured AirCore data can be used to contribute to the TCCON calibration (Wunch et al., 2010).

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3

1 Table 1. Measurement settings for the Sodankylä Bruker 125HR FTS instrument.

Item	Setting
Aperture	1.0 mm
Detectors	RT-Si Diode DC + RT-InGaAs DC
Scanner velocity	10 kHz
Low Pass Filter	10 kHz
High Folding Limit	15798.007031
Resolution	0.020000
Acquisition Mode	Single Sided, Forward-Backward
Sample Scans	2

2

3

- 1 Table 2. Laser board settings and measurements. Ghost to parent intensity ratio GPR and the ratio of the spurious signal to primary signal
2 intensity SPR are shown at different scanner velocities. Values used for our measurements are shown in bold.

Period	Laser board	Laser detectors	Pressure hPa	Ghost minimized kHz	Filter wavenumber cm-1	Velocity kHz	GPR(4150 cm ⁻¹) 10 ⁻⁴	SPR 10 ⁻⁴
up to 9.3.2010	ECL02	V01	0.4	-	4315	5	7.4	8.1
						10	13	8.2
						20	26	7.7
10.3.2010 to 2.3.2011	ECL04	V01	0.16	10	5960	7.5	0.42	8.0
						10	0.75	8.1
						20	8.2	8.1
						40	33	8.0
3.3.2011 to present	ECL05	V02	0.7	10	5960	7.5	0.19	8.3
						10	0.14	8.4
						20	0.56	8.4
						40	1.3	8.3

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4

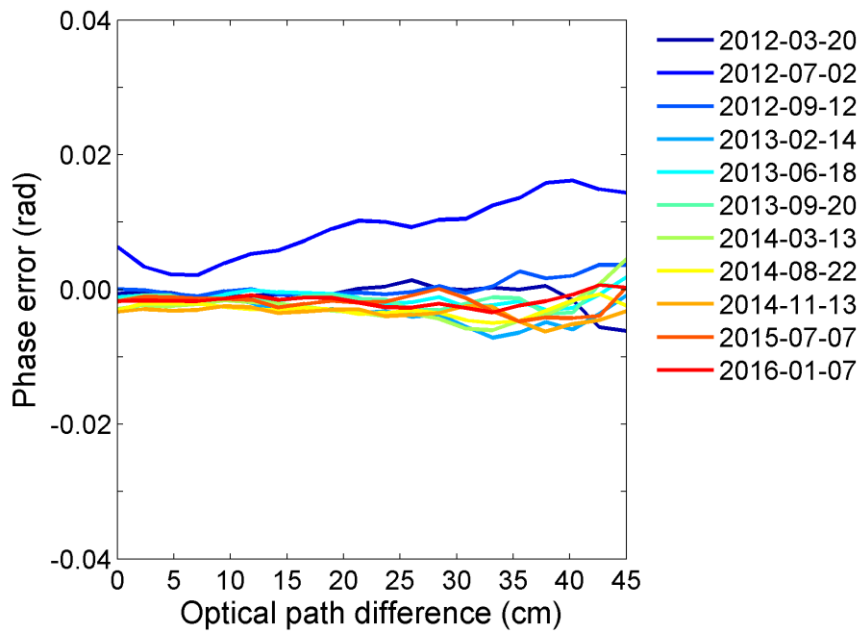
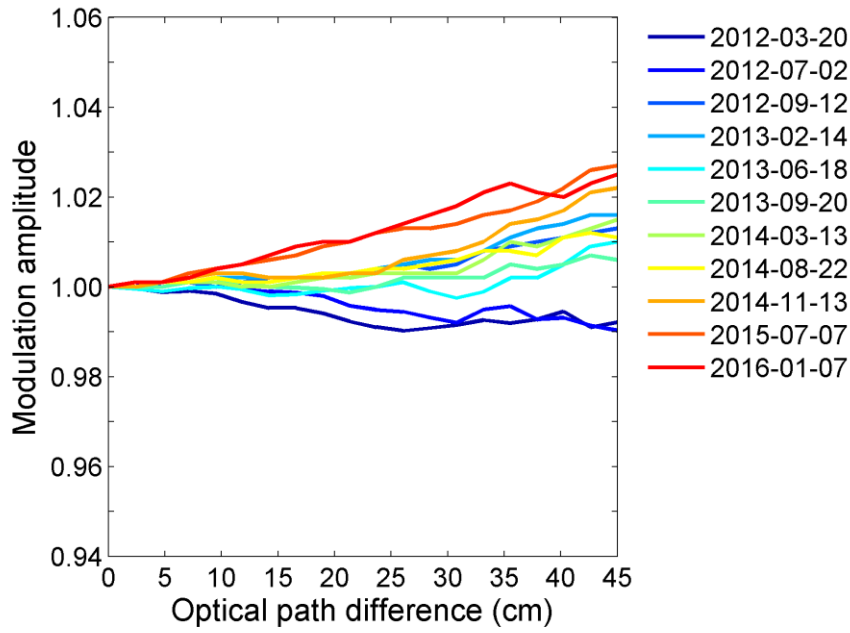
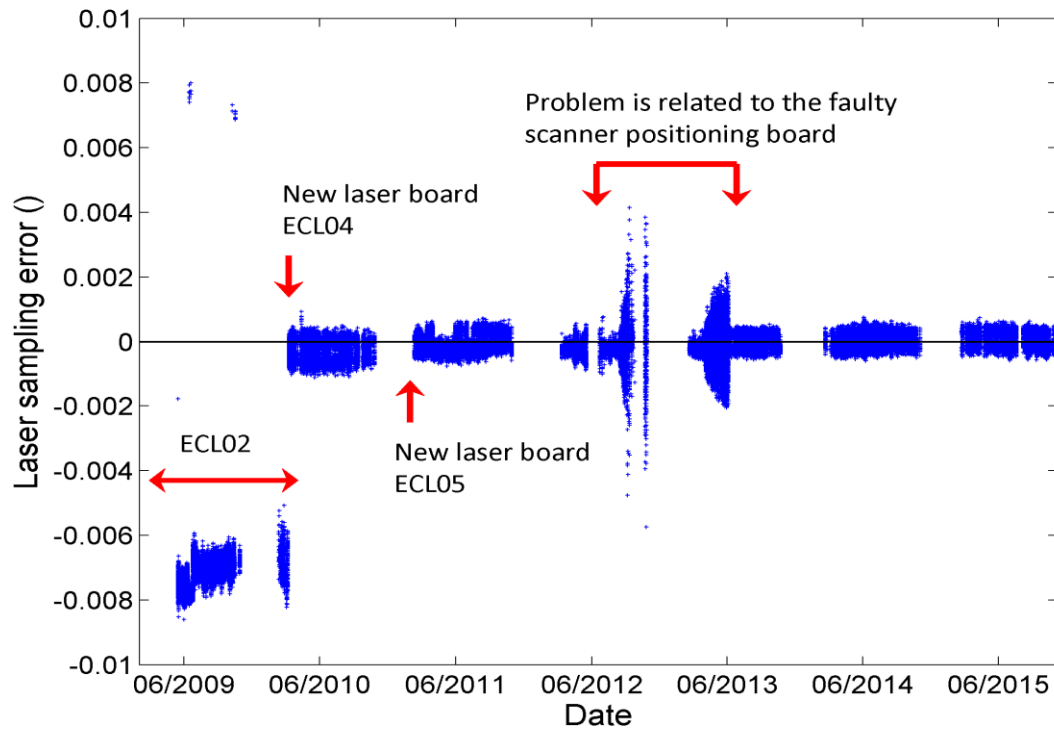


Figure 1. Time series of measurements of modulation efficiency: amplitude (upper panel) and phase errors (lower panel) are shown as a function of optical path difference.



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3 Figure 2. Laser sampling error (LSE) since 2009. LSE correction is applied during the
 4 retrieval process within GGG2014.

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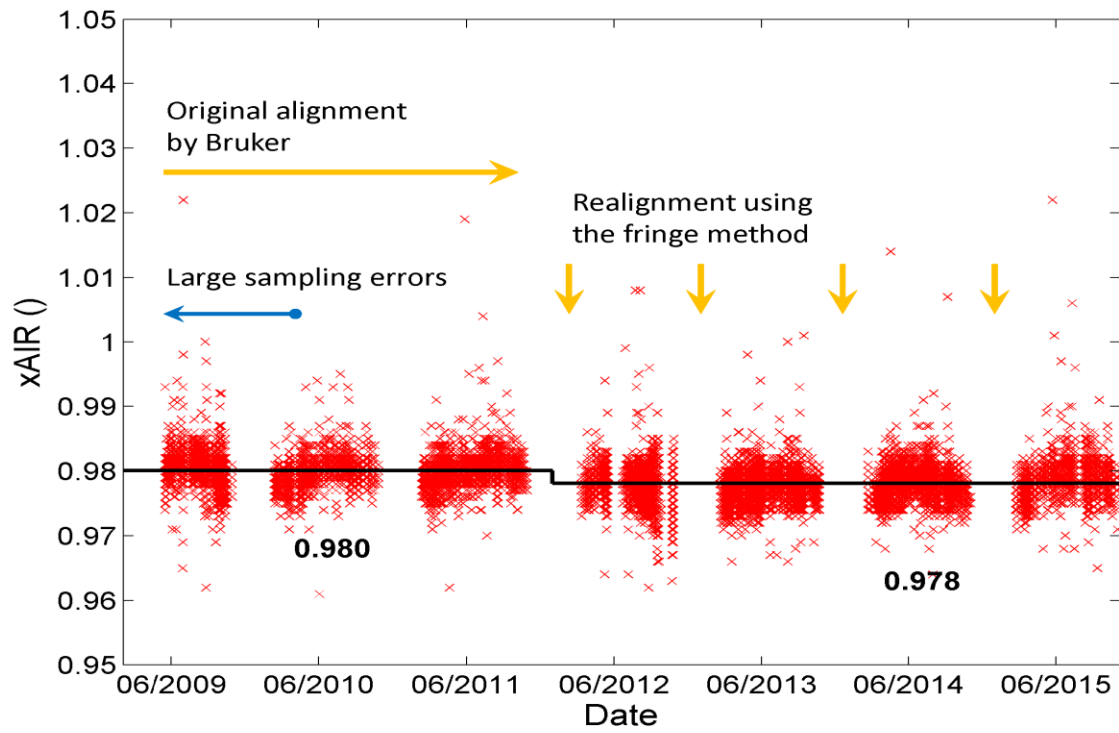


Figure 3. Time series of xAIR. Average xAIR values are shown for 2009-2011 (0.980) and for 2012-2015 (0.978).

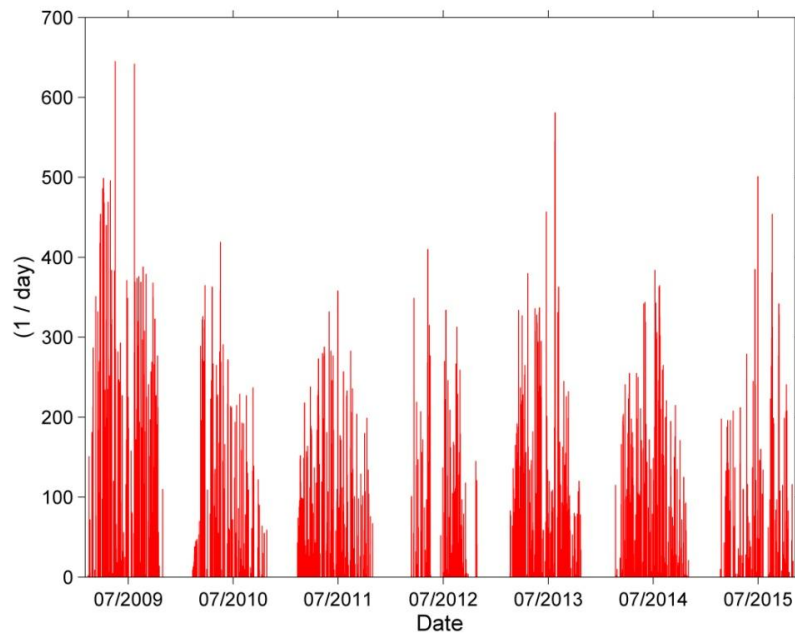


Figure 4. Distribution of FTS measurements per day at Sodankylä during 2009-2015. Criteria for an accepted measurement shown here is solar zenith angle $< 82^\circ$ and solar intensity variation $< 5\%$. In total 123715 spectra were recorded during the 7 year period, corresponding to 1022 measurement days.

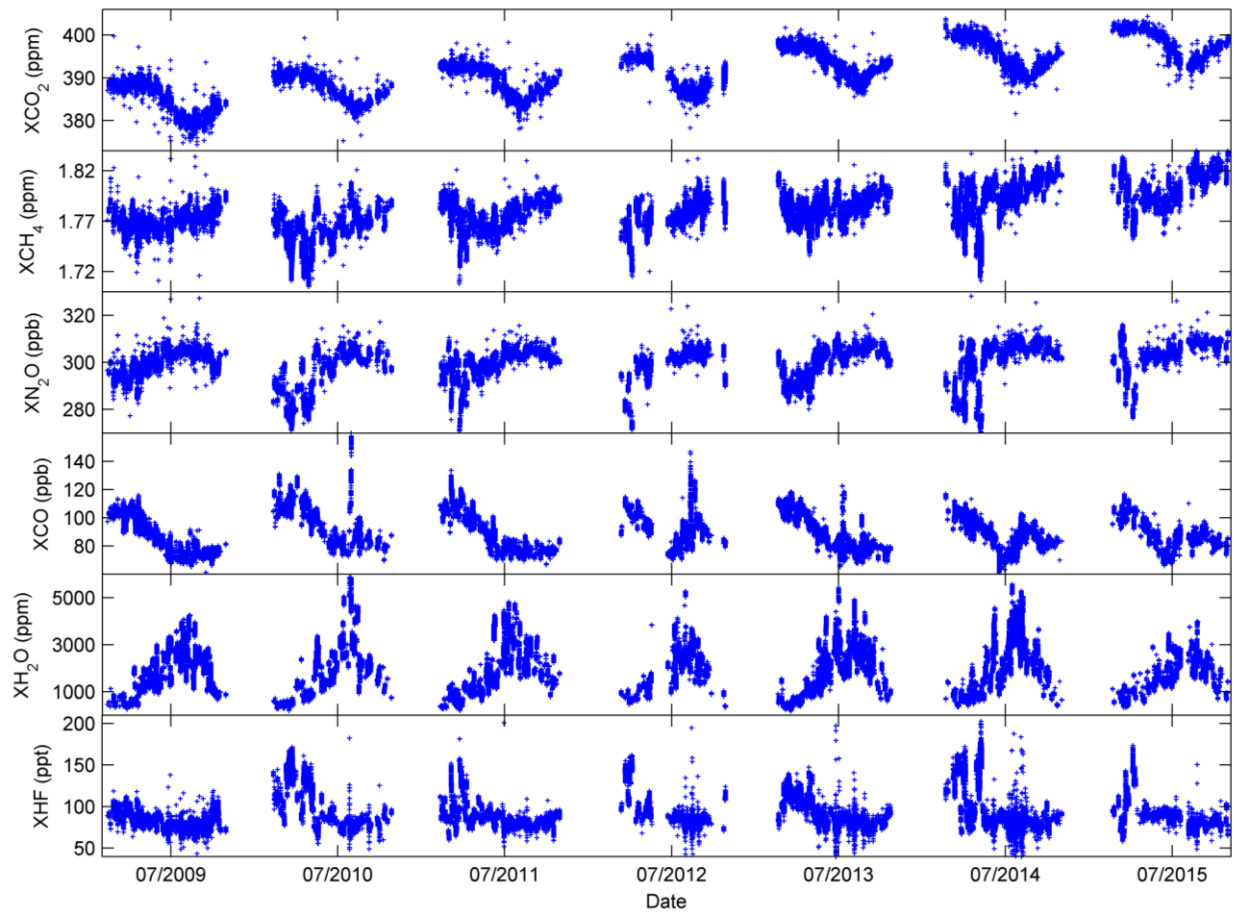
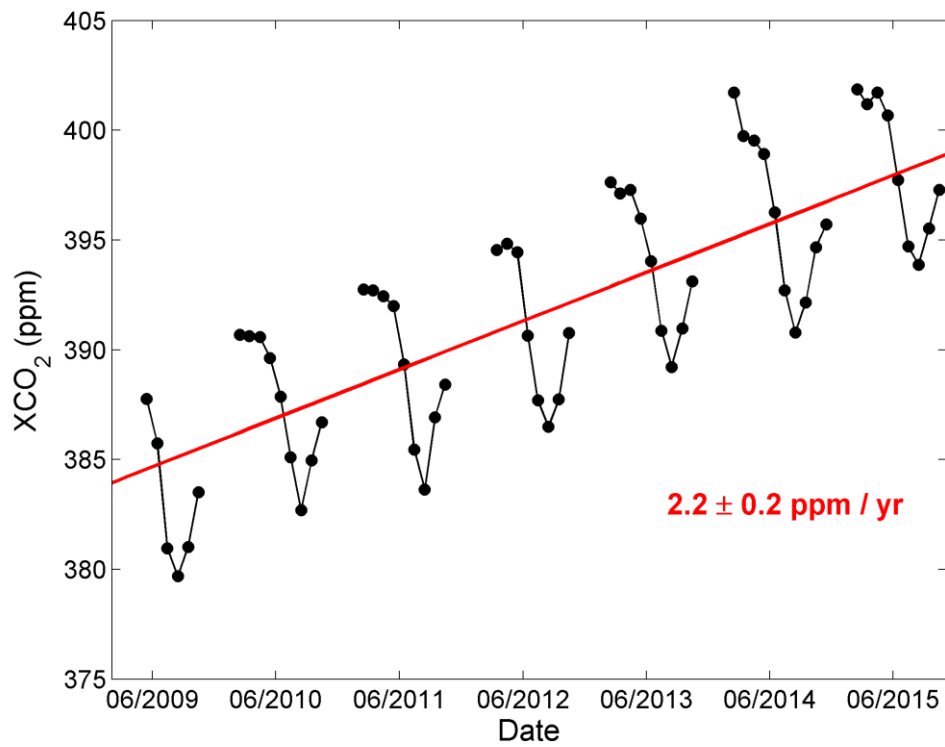


Figure 5. Time series of XCO₂ measurements at Sodankylä since May 2009 (upper panel). Each marker indicates a single measurement. Lower panels correspond to other gases retrieved from the same measurements.

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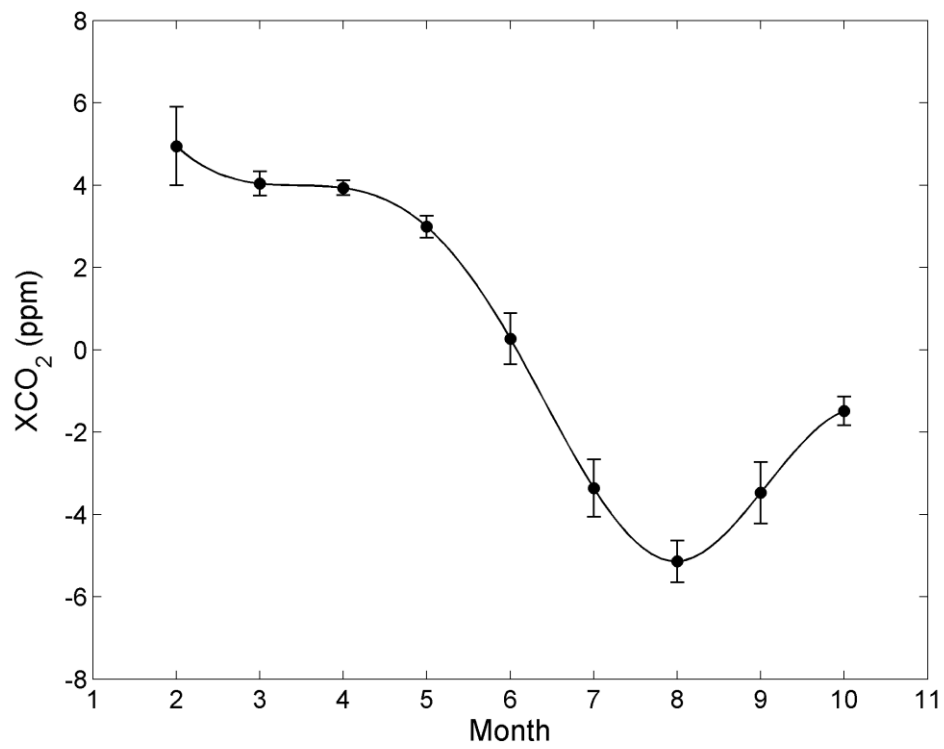


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4 Figure 6. Time series of XCO₂ measurements at Sodankylä since May 2009. Each marker
5 indicates monthly mean. A trend of 2.2+/-0.2 ppm per year has been observed during 2009-
6 2015.

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4 Figure 7. Average seasonal cycle of XCO₂ over Sodankylä, monthly averages (black dots)
5 and standard deviations (vertical lines). The average seasonal cycle was calculated after the
6 trend removal.

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