Fourier transform spectrometer measurements of column CO$_2$ at Sodankylä, Finland

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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 CO$_2$ time series over a 7-year period (2009–2015) and provide a description of the FTS system and data processing at Sodankylä. We find the lowest monthly column CO$_2$ values in August and the highest monthly values during the February–May season. Inter-annual variability is the highest in the June–September period, which correlates with the growing season. During the time period of FTS measurements from 2009 to 2015, we have observed a 2.2 ± 0.2 ppm increase per year in column CO$_2$. The monthly mean column CO$_2$ values have exceeded 400 ppm level for the first time in February 2014.

1 Introduction
Carbon dioxide (CO$_2$) is the most abundant anthropogenic greenhouse gas in the atmosphere (Hartman et al., 2013). The concentration of CO$_2$ has increased rapidly since the industrial revolution due to the burning of carbon-based fuels. Precise and accurate measurements of CO$_2$ are needed in order to better understand the carbon cycle. In addition to the relatively long term in situ measurements of CO$_2$, ground-based total column measurements of carbon dioxide have become possible more recently. The column-averaged, dry-air mole fractions of carbon dioxide (XCO$_2$) have been measured since the year 2004 by the Total Carbon Column Observing Network (TCCON) sites, using solar Fourier transform spectrometers (FTSs), 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 XCO$_2$, but also other gases have been retrieved, including CH$_4$, CO, N$_2$O, H$_2$O, HDO and HF. Compared to the surface in situ measurements, XCO$_2$ is less affected by changes in the height of the planetary boundary layer and the spatial sensitivity footprint is larger (Keppel-Aleks et al., 2011). The accuracy and precision of the XCO$_2$ measurements within TCCON are better than 0.25 % (Wunch et al., 2011a). The high accuracy and precision are needed to contribute to the carbon cycle research and validation of spaceborne 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 XCO$_2$ retrievals have been used in several studies (e.g., 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 re-
trieval version GGG2014 (Wunch et al., 2015). The quality-controlled data from May 2009 to November 2015 have been used here to calculate the average seasonal cycle and trend of XCO$_2$ over the measurement period.

2 Instrumentation

The Sodankylä TCCON FTS station is part of the infrastructure of the Finnish Meteorological Institute’s Arctic Research Centre. The station is located at 67.3668° N, 26.6310° E, 188 m.a.s.l. FTS measurements at Sodankylä are made using a Bruker 125HR 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, manufactured by Bruker Optics. The cover of the tracker was built locally at the institute’s workshop.

The FTS instrument is equipped with two room-temperature detectors: an indium gallium arsenide (InGaAs, covering 4000–11 000 cm$^{-1}$) and a silicon diode (Si, covering 9000–15 000 cm$^{-1}$), which is similar to the other FTS stations in the TCCON network. The measurements are performed in a vacuum to improve stability and to reduce water vapor in the system. The system is evacuated each night to avoid vibration during the solar measurements. The optical path difference (OPD) is 45 cm and the spectral resolution is 0.02 cm$^{-1}$; collection time for a single scan is 78 s. Column abundances of CO$_2$, O$_2$, CH$_4$, H$_2$O, HDO, HF, CO and N$_2$O are retrieved from the spectra.

The FTS instrument has worked in a fully automated mode since July 2013. Readings from rain and direct solar radiation sensors, combined with the automated analysis of weather radar forecast data, determine the start and cessation of daily measurements. A control system monitors the measurement quality and automatically reports on error conditions, thus longer measurement gaps have been minimized. Currently used settings are presented in Table 1. In addition to the TCCON measurements, we also take longer wavelength measurements, using a liquid nitrogen cooled indium antimonide detector (InSb, covering 1800–6000 cm$^{-1}$). The InSb measurements are filtered, the pass band is at 2439–3125 cm$^{-1}$. This filter choice is designed for profile retrievals of methane and provides a possibility to compare the mid-infrared and near-infrared retrievals of CH$_4$. The sequence of measurements is such that after two InGaAs/Si scans, one InSb scan is taken. To be able to make the solar intensity variation correction, we have recorded all interferograms in the DC mode.

To guarantee the optimal performance of the instrument, the optical alignment is checked and adjusted at least once per year. Usually the alignment is performed in winter, be-cause then the solar measurements are not possible due to the high-latitude location of the station. We have applied the alignment procedure developed by Hase and Blumenstock (2001). The alignment method is based on the inspection of laser fringes through a telescope. In addition we monitor the instrument line shape (ILS) by taking HCl reference gas measurements on a monthly basis. The ILS retrievals are made using the LINEFIT14 software (Hase et al., 2013). Figure 1 presents a selection of ILS retrievals. The upper panel corresponds to the amplitude of the modulation and the lower panel to the phase error, both as functions of optical path 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 the case of Sodankylä, 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 (Fig. 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, XCO$_2$ values are retrieved in two bands, centered at 6228 and 6348 cm$^{-1}$. Within TCCON, the retrieval of XCO$_2$ 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 a slightly different setup of instrumentation. For example, not all the TCCON stations have the Si detector available.

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

<table>
<thead>
<tr>
<th>Item</th>
<th>Setting</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aperture</td>
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<tr>
<td>Detectors</td>
<td>RT-Si Diode DC, RT-InGaAs DC</td>
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<tr>
<td>Scanner velocity</td>
<td>10 kHz</td>
</tr>
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<td>Low pass filter</td>
<td>10 kHz</td>
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<tr>
<td>High folding limit</td>
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</tr>
<tr>
<td>Resolution</td>
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</tr>
<tr>
<td>Acquisition mode</td>
<td>Single sided, forward–backward</td>
</tr>
<tr>
<td>Sample scans</td>
<td>2</td>
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</tbody>
</table>

Table 2. Laser board settings and measurements. Ghost-to-parent intensity ratio (GPR) and the ratio of the spurious signal to primary signal intensity (SPR) are shown at different scanner velocities. The used scanner velocities and the corresponding GPR and SPR values are shown in bold.

<table>
<thead>
<tr>
<th>Period</th>
<th>Laser board</th>
<th>Laser detectors</th>
<th>Pressure</th>
<th>Ghost minimized wavenumber</th>
<th>Filter</th>
<th>Velocity</th>
<th>GPR (4150 cm(^{-1}))</th>
<th>SPR (10^{-4})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Up to 9 Mar 2010</td>
<td>ECL02</td>
<td>V01</td>
<td>0.4</td>
<td>–</td>
<td>4315</td>
<td>5</td>
<td>7.4</td>
<td>8.1</td>
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<td></td>
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<td></td>
<td>10</td>
<td>13</td>
<td>8.2</td>
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<td></td>
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<td></td>
<td></td>
<td>20</td>
<td>26</td>
<td>7.7</td>
</tr>
<tr>
<td>10 Mar 2010 to 2 Mar 2011</td>
<td>ECL04</td>
<td>V01</td>
<td>0.16</td>
<td>10</td>
<td>5960</td>
<td>7.5</td>
<td>0.42</td>
<td>8.0</td>
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<td></td>
<td>10</td>
<td>0.75</td>
<td>8.1</td>
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<td>8.2</td>
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<td>40</td>
<td>33</td>
<td>8.0</td>
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<tr>
<td>3 Mar 2011 to present</td>
<td>ECL05</td>
<td>V02</td>
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<td>10</td>
<td>5960</td>
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<td>40</td>
<td>1.3</td>
<td>8.3</td>
</tr>
</tbody>
</table>

XCO\(_2\), the column-averaged, dry-air mole fraction of CO\(_2\), is defined as the ratio of CO\(_2\) 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 O\(_2\). The oxygen column is retrieved from the TCCON FTS spectra and the method via oxygen is adopted in TCCON. XCO\(_2\) is the ratio of CO\(_2\) column to O\(_2\) column,

\[
XCO_2 = \frac{\text{CO}_2 \text{column}}{\text{O}_2 \text{column}} \times 0.2095. \tag{1}
\]

By calculating the ratio, all errors that affect both columns cancel in the same way. This improves the repeatability of the XCO\(_2\) 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 3 March 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 the case of our instrument. The ECL02 board was installed on 10 March 2010, and was replaced a year later (Table 2). The currently used electronic board (ECL05) has been operational since 3 March 2011. Intermittent fluctuations in LSE from 27 August to 11 November 2012 and again from 6 July to 1 August 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 2 August 2013 and since then the sampling errors have been minimal.

Another important measure of data quality and instrument performance is xAIR, the column-averaged, 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\(_2\)O, to the total dry air column, obtained from the measured oxygen column:

\[
xAIR = \frac{\text{AIR column}}{\text{O}_2 \text{ column}} \times 0.2095 - \text{XH}_2\text{O} \times \frac{m_{\text{H}_2\text{O}}}{m_{\text{air}}} \tag{2}
\]

\[
x\text{AIR column} = \frac{P_S}{\frac{m_{\text{air}}}{N_A}} \times \frac{m_{\text{dry}}}{m_{\text{air}}} \tag{3}
\]

\(m_{\text{H}_2\text{O}}\) and \(m_{\text{air}}\) are the molecular masses of water vapor and dry air, \(N_A\) is Avogadro’s constant and \(\{g\}_{\text{air}}\) 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 (Washenfelder 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 Fig. 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).

The xAIR record shows that the instrument has been stable during its history. xAIR behaves consistently also during the period of relatively large sampling errors, because of the resampling included in the GGG2014 processing scheme. This was not the case with the previous version of data reprocessing system, GGG2012. In the previous data version, the xAIR level was too low for the given period of measurements. During the first months of year 2009 we did not have a dichroic beamsplitter installed and therefore we had no Si measurements. Reprocessing the earliest data, from the time period 6 February 2009–15 May 2009 needs a different approach (Dohe et al., 2013). Therefore, the data from this time period have not been reprocessed using GGG2014. For the previous data version (GGG2012) we have made an additive LSE correction for the given time period, based on the data collected at different scanner speeds. Without any LSE correction, the xGAS values are too low for these months by amounts ranging from 0.2 to 1.0 %. The calculated additive correction for XCO₂ is 2.5 ppm. For other gases, the correction is as follows: XCO 0.86 ppb, XCH₄ 0.012 ppm, XH₂O 2.9 ppm and XN₂O 2.4 ppb.
The GGG2014 data version in this study covers the time period from 15 May 2009 to 5 November 2015. During these years we have collected 111,825 individual measurements, which have been spread over 966 days. In addition, 11,890 measurements were made over 56 days during 16 February–15 May 2009, which were included in data version GGG2012. Thus the total number of measurements has been 123,715 over 1022 days (Fig. 4). A single measurement was graded as acceptable if the solar intensity variation during the measurement was less than 5% and the solar zenith angle was less than 82°. Due to the zenith angle constraint, good measurements are only possible from 8 February to 11 November each year (268 days), resulting in a gap in winter that is over 3 months long. On average, there have been 146 measurement days per year. The main factor that limits the amount of measurements is cloudiness, though measurement gaps also occur due to technical problems. A 1-month gap in the measurements was caused by the failure of sampling laser on 20 May 2012; the laser was replaced on 20 June 2012. A slight increase in the amount of measurements can be observed in 2013 because this was the first year when the instrument worked in fully automatic mode.

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

4 XCO\textsubscript{2} time series and the annual cycle

The XCO\textsubscript{2} measurements are presented in Fig. 5 (upper panel), corresponding to the time period of 2009–2015. All available data are shown, including version GGG2012 data until 15 May 2009 and the proceeding data retrieval version GGG2014 data. We have also included time series of other gases that are retrieved together with the XCO\textsubscript{2}. The other time series are for XCH\textsubscript{4}, XN\textsubscript{2}O, XCO, XH\textsubscript{2}O and XHF measurements. The non-CO\textsubscript{2} TCCON measurements from Sodankylä have been previously published by, e.g., Saito et al. (2012); Belikov et al. (2013); Mielonen et al. (2013); Yoshida et al. (2013); Saad et al. (2014); Tsuruta et al. (2015); Dupuy et al. (2016); Inoue et al. (2016).

Over the 7-year time period, the trend of XCO\textsubscript{2} is found to be $2.2 \pm 0.2$ ppm yr\textsuperscript{−1} ($\pm 1$ standard error). In Fig. 6, monthly mean values are plotted for each month when measurements have been possible. GGG2014 data version has been used for the trend calculation. 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 XCO\textsubscript{2} 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 XCO\textsubscript{2}, we find a significant trend in XCH\textsubscript{4}. In the case of XCH\textsubscript{4}, the observed increase has been $7.1 \pm 0.8$ ppb yr\textsuperscript{−1}.

The average annual cycle of XCO\textsubscript{2} is shown in Fig. 7, based on the 7 years of measurement and the GGG2014 retrieval. The highest values of XCO\textsubscript{2} are obtained in February to May period, before the start of the growing season. The minimum monthly XCO\textsubscript{2} occurs in August due to the uptake of carbon into the biosphere, which correlates with the period of plant growth. The inter-annual variability is found to be the smallest in spring (March–May) and largest in summer and autumn (June–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 the XCO\textsubscript{2} annual cycle is correlated with summertime surface temperature anomalies. The amplitude of the column CO\textsubscript{2} seasonal cycle at high latitudes of the Northern Hemisphere is smaller than the one based on surface measurement (Olsen and Randerson, 2004). Column CO\textsubscript{2} 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 XCO\textsubscript{2}. 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 CO\textsubscript{2} have also become available through Monitoring of Atmospheric Composition and Climate – Interim.
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 \(2.2 \pm 0.2\) ppm increase per year in \(XCO_2\) values. In February 2014 the monthly mean \(XCO_2\) values have exceeded 400 ppm level for the first time in the history of these measurements. The lowest monthly \(XCO_2\) 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 \(CO_2\), \(CH_4\) 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-round measurements by AirCore are possible. The AirCore used in Sodankylä is a 100 m long coiled sampling tube, with a volume of \(\approx 1400\) ml (Paul et al., 2016). The sampling tube is filled during the payload descent and is automatically closed 9 s after the landing. Gas analysis have been performed by a cavity ring-down spectrometer (Picarro Inc., CA, model G2401), typically with a start of the analysis within 2–3 h after each AirCore flight. 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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References


