

1 **FTS measurements of column CO<sub>2</sub> at Sodankylä**

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6

7 **Abstract**

8 Fourier Transform Spectrometer (FTS) observations at Sodankylä, Finland (67.4° N, 26.6 ° E)  
9 have been performed since early 2009. The FTS instrument is participating in the Total  
10 Carbon Column Observing Network (TCCON) and has been optimized to measure  
11 abundances of the key greenhouse gases in the atmosphere. Sodankylä is the only TCCON  
12 station in the Fennoscandia region. Here we report the measured CO<sub>2</sub> time series over a seven  
13 year period (2009-2015) and provide a description of the FTS system and data processing at  
14 Sodankylä. We find the lowest monthly column CO<sub>2</sub> values in August and the highest  
15 monthly values during the February to May season. Inter-annual variability is the highest in  
16 June-September period, which correlates with the growing season. During the time period of  
17 FTS measurements from 2009 until 2015 we have observed a 2.2+-0.2 ppm increase per year  
18 in column CO<sub>2</sub>. The monthly mean column CO<sub>2</sub> values have exceeded 400 ppm level for the  
19 first time in February 2014.

20

21 **1 Introduction**

22 Carbon dioxide (CO<sub>2</sub>) is the most abundant anthropogenic greenhouse gas in the atmosphere  
23 (Hartman et al., 2013). The concentration of CO<sub>2</sub> has increased due to the burning of carbon-  
24 based fuels. Precise and accurate measurements of CO<sub>2</sub> are needed in order to better  
25 understand the carbon cycle. In addition to the relatively long term *in situ* measurements of  
26 CO<sub>2</sub>, ground based total column measurements of carbon dioxide have become possible more  
27 recently. The column averaged dry-air mole fractions of carbon dioxide (XCO<sub>2</sub>) have been  
28 measured since year 2004 by the Total Carbon Column Observing Network (TCCON) sites,  
29 using solar Fourier Transform Spectrometers (FTS), operating in the near infrared spectral

1 region (Wunch et al., 2011a). The main goal of the TCCON has been to provide precise and  
2 accurate measurements of XCO<sub>2</sub>, but also other gases have been retrieved, including CH<sub>4</sub>,  
3 CO, N<sub>2</sub>O, H<sub>2</sub>O, HDO and HF. Compared to the surface *in situ* measurements XCO<sub>2</sub> is less  
4 affected by changes in the height of planetary boundary layer and the spatial sensitivity  
5 footprint is larger (Keppel-Aleks et al., 2011).. The accuracy and precision of the XCO<sub>2</sub>  
6 measurements within TCCON is better than 0.25% (Wunch et al., 2011a). The high accuracy  
7 and precision is needed to contribute to the carbon cycle research and validation of space  
8 borne measurements. Satellite missions that have already used the TCCON data include the  
9 Orbiting Carbon Observatory-2 (OCO-2; Crisp et al., 2004); the Greenhouse Gases Observing  
10 Satellite (GOSAT; Yokota et al., 2009) and the SCanning Imaging Absorption SpectroMeter  
11 for Atmospheric CHartographY (SCIAMACHY; Bovensmann et al., 1999).

12 Sodankylä in northern Finland is one of the stations in the TCCON. This is currently the only  
13 TCCON station in the Fennoscandia region. We established the FTS measurements at  
14 Sodankylä in early 2009. Since then the XCO<sub>2</sub> retrievals have been used in several studies  
15 (e.g. in Wunch et al., 2011b; Oshchepkov et al., 2012; Saito et al., 2012; Belikov et al., 2013;  
16 Guerlet et al., 2013; Yoshida et al., 2013; Agustí-Panareda, 2014; Deng et al., 2014; Reuter et  
17 al., 2014; Barthlott et al., 2015; Heymann et al., 2015; Lindqvist et al., 2015; Belikov et al.,  
18 2016; Feng et al., 2016; Inoue et al., 2016; Massart et al., 2016). This paper describes the  
19 instrumentation, measurement procedures and data processing at the Sodankylä FTS site,  
20 corresponding to the data retrieval version GGG2014 (Wunch et al., 2015). The quality  
21 controlled data from May 2009 until November 2015 have been used here to calculate the  
22 average seasonal cycle and trend of XCO<sub>2</sub> over the measurement period.

23

## 24 **2 Instrumentation**

25 The Sodankylä TCCON FTS station is part of the infrastructure of the Finnish Meteorological  
26 Institute's Arctic Research Center. The FTS is located at 67.3668° N, 26.6310 ° E, 188 m.a.s.  
27 FTS measurements at Sodankylä are made using a Bruker 125 HR FTS (Bruker Optics,  
28 Germany). Since the beginning of the data record the FTS instrument has been installed in a  
29 two-story observational building. The interior of the laboratory was rebuilt in late 2008 to  
30 mount the FTS instrument. The instrument is placed on a concrete plate, which is designed to  
31 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  $\text{cm}^{-1}$ ) and a silicon diode (Si, covers 9000-15000  $\text{cm}^{-1}$ ),  
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 optical  
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  $\text{cm}^{-1}$ , collection time for a  
9 single scan is 78 seconds.. Column abundances of CO<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O, HDO, HF, CO and  
10 N<sub>2</sub>O 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  $\text{cm}^{-1}$ ). The InSb  
18 measurements are filtered, the pass-band is at 2439-3125  $\text{cm}^{-1}$ . 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<sub>4</sub>. 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

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

11

### 12 **3 Data processing and availability**

13 Using the InGaAs detector,  $\text{XCO}_2$  values are retrieved in two bands, centered at  $6228 \text{ cm}^{-1}$   
14 and  $6348 \text{ cm}^{-1}$ . Within TCCON, the retrieval of  $\text{XCO}_2$  and other gases is based on the GFIT  
15 algorithm as described by Wunch et al. (2011a). The data processing and analysis scheme is  
16 common at each TCCON site, although some sites may have slightly different setup of  
17 instrumentation. For example, not all the TCCON stations have the Si detector available.

18  $\text{XCO}_2$ , the column-averaged dry-air mole fraction of  $\text{CO}_2$ , is defined as the ratio of  $\text{CO}_2$  total  
19 column to the total column of all gases, excluding water. The total dry air column can be  
20 calculated either from surface pressure and water vapor column or from oxygen column,  
21 assuming the constant dry-air mole fraction of 20.95% for  $\text{O}_2$ . The oxygen column is retrieved  
22 from the TCCON FTS spectra and the method via oxygen is adopted in TCCON.  $\text{XCO}_2$  is the  
23 ratio of  $\text{CO}_2$  column to  $\text{O}_2$  column,

$$24 \quad \text{XCO}_2 = \frac{\text{CO}_2 \text{ column}}{\text{O}_2 \text{ column}} \times 0.2095 \quad (1)$$

25 By calculating the ratio, all errors that affect both columns in the same way cancel. This  
26 improves the repeatability of the  $\text{XCO}_2$  retrieval.

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

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

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

$$16 \quad xAIR = \frac{AIR\ column}{O_2\ column} \times 0.2095 - XH_2O \times \frac{m_{H_2O}}{m_{air}^{dry}} \quad (2)$$

$$17 \quad AIR\ column = \frac{P_S}{\{g\}_{air} \times \frac{m_{air}^{dry}}{N_A}} \quad (3)$$

18  $m_{H_2O}$  and  $m_{air}^{dry}$  are the molecular masses of water vapor and dry air,  $N_A$  is Avogadro's  
 19 constant and  $\{g\}_{air}$  is the column-averaged gravitational acceleration. Ideally this ratio should  
 20 be 1, but typically the xAIR value is little less, around 0.98, in TCCON measurements, related  
 21 to errors in the O<sub>2</sub> spectroscopy (Washenfelder et al., 2006). In practice xAIR is a measure of  
 22 how well the instrument is capable of obtaining the oxygen column. Large differences in  
 23 xAIR values compared to the network wide mean are a sign of instrument problems. The  
 24 problems may be related to several factors, such as a poor optical alignment, spectral ghosts  
 25 or faulty pressure sensor.

26 The time series of xAIR are shown in Figure 3. The average xAIR value for 2009-2011 is  
 27 0.980 and the average xAir for the time period of 2012-2015 is 0.978. The first 3 years, until  
 28 2012, correspond to the original alignment by Bruker, while the realignment since 2012 was  
 29 performed using the fringe method. The method is considered an improvement over the  
 30 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 XCO<sub>2</sub> is 2.5 ppm.  
13 For other gases the correction is as follows: XCO 0.86 ppb, XCH<sub>4</sub> 0.012 ppm, XH<sub>2</sub>O 2.9 ppm  
14 and XN<sub>2</sub>O 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

1    **4 XCO<sub>2</sub> time series and the annual cycle**

2    The absolute values of each of the XCO<sub>2</sub> measurements are presented in Figure 5 (upper  
3    panel), corresponding to the time period of 2009-2015. We have also included time series of  
4    other gases that are retrieved together with the XCO<sub>2</sub>, using GGG2014. The other time series  
5    are for XCH<sub>4</sub>, XN<sub>2</sub>O, XCO, XH<sub>2</sub>O and XHF measurements. The non-CO<sub>2</sub> TCCON  
6    measurements from Sodankylä have been previously published by e.g. Saito et al. (2012);  
7    Belikov et al. (2013); Yoshida et al. (2013); Saad et al. (2014); Tsuruta et al. (2015); Inoue et  
8    al. (2016).

9    Over the seven year time period the trend of XCO<sub>2</sub> is found to be 2.2+/-0.2 ppm/year (+/- one  
10   standard error). In Figure 6 monthly mean values are plotted for each month when  
11   measurements have been possible. The trend is in broad agreement with earlier studies (e.g.  
12   Lindqvist et al., 2015), though it is based on a longer time period. It is noteworthy that in  
13   February 2014 the monthly mean XCO<sub>2</sub> values have 400 ppm level for the first time, while  
14   individual measurements have achieved the 400 ppm level already in spring 2012 and 2013.  
15   Similar to the XCO<sub>2</sub>, we find significant trend in XCH<sub>4</sub>. In case of XCH<sub>4</sub> the observed  
16   increase has been 7.1 +/- 0.8 ppb/year.

17   The average annual cycle of XCO<sub>2</sub> is shown in Figure 7, based on 7 years of measurement.  
18   The highest values of XCO<sub>2</sub> are obtained in February to May period, before the start of the  
19   growing season. The minimum monthly XCO<sub>2</sub> occurs in August due to the uptake of carbon  
20   into the biosphere, which correlates with the period of plant growth. The interannual  
21   variability is found to be the smallest in spring (March-May) and largest in summer and  
22   autumn (June to September). The shape of the annual cycle can be explained by the imbalance  
23   between ecosystem respiration and gross primary production. This is often referred to as net  
24   ecosystem exchange (NEE). At high latitudes a negative NEE is observed during the growing  
25   season, because the gross primary production has a peak around the summer solstice, while  
26   ecosystem respiration has a maximum later in summer, in correlation with the increase in  
27   ground and air temperature (Lloyd and Taylor, 1994). Based on the TCCON measurements,  
28   Wunch et al. (2013) found that the minima in XCO<sub>2</sub> annual cycle is correlated with  
29   summertime surface temperature anomalies. The amplitude of the column CO<sub>2</sub> seasonal cycle  
30   at high latitudes of the Northern Hemisphere is smaller than the one based on surface  
31   measurement (Olsen and Randerson, 2004). Column CO<sub>2</sub> seasonal variability can be  
32   explained by the variability in the terrestrial biospheric fluxes (Keppel-Aleks et al., 2011),

1 while the long-term trend is driven by the fossil fuel emissions (Hartman et al., 2013).  
2 CarbonTracker (Peters et al., 2007) has been widely used to study the annual cycle of XCO<sub>2</sub>.  
3 It has been shown that CarbonTracker is able to simulate the seasonal cycle at Sodankylä with  
4 an average model-measurement bias less than 0.4 ppm (Reuter et al., 2014). Recently the  
5 daily forecasts of CO<sub>2</sub> have also become available through Monitoring of Atmospheric  
6 Composition and Climate - Interim Implementation (MACC-II) service at the European  
7 Centre for Medium- Range Weather Forecasts. The model includes also the short term  
8 meteorological variability (Agustí-Panareda et al., 2014).

9

## 10 **5 Conclusions and outlook**

11 XCO<sub>2</sub> measurements have been made at Sodankylä since early 2009. The FTS instrument has  
12 been relatively stable. Regular instrument alignments and HCl cell measurements have been  
13 performed. The instrument has run in fully automatic mode since 2013, therefore the temporal  
14 data coverage is relatively good, given the high latitude conditions at Sodankylä. The  
15 historical data have been reprocessed using the GGG2014 software (Wunch et al., 2015). The  
16 data have been made available via the Carbon Dioxide Information Analysis Center, Oak  
17 Ridge National Laboratory, Oak Ridge, Tennessee, USA (Kivi et al., 2014). Measurements  
18 from other TCCON sites are also available from the same data center.

19 Based on the measurements at Sodankylä we find a 2.2+/-0.2 ppm increase per year in XCO<sub>2</sub>  
20 values. In February 2014 the monthly mean XCO<sub>2</sub> values have exceeded 400 ppm level for  
21 the first time in the history of these measurements. The lowest monthly XCO<sub>2</sub> values within  
22 the seasonal cycle are found in August and the highest in February-May. Year-to-year  
23 variability is lowest is March-May and highest during the growing season in June-September.

24 Relevant to the FTS measurements, we have started with balloon borne AirCore (Karion et  
25 al., 2010) profile measurements of CO<sub>2</sub>, CH<sub>4</sub> and CO at Sodankylä in September 2013. The  
26 balloon measurements have the benefit of reaching much higher vertical altitudes (up to 30-35  
27 km), compared to the aircraft *in situ* measurements. In addition, year around measurements by  
28 AirCore are possible. AirCore used in Sodankylä is a 100 m long coiled sampling tube, with a  
29 volume of  $\approx$  1400 ml (Paul et al., 2016). The sampling tube is filled during the payload  
30 descent and is automatically closed 9 seconds after the landing. The profile analysis has been  
31 performed typically within 2-3 hours after the landing of the payload. Gas analysis have been  
32 performed by a Cavity Ring-Down Spectrometer (Picarro Inc., CA, model G2401). Total gas

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

4

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8

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3

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

<b>Item</b>	<b>Setting</b>
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

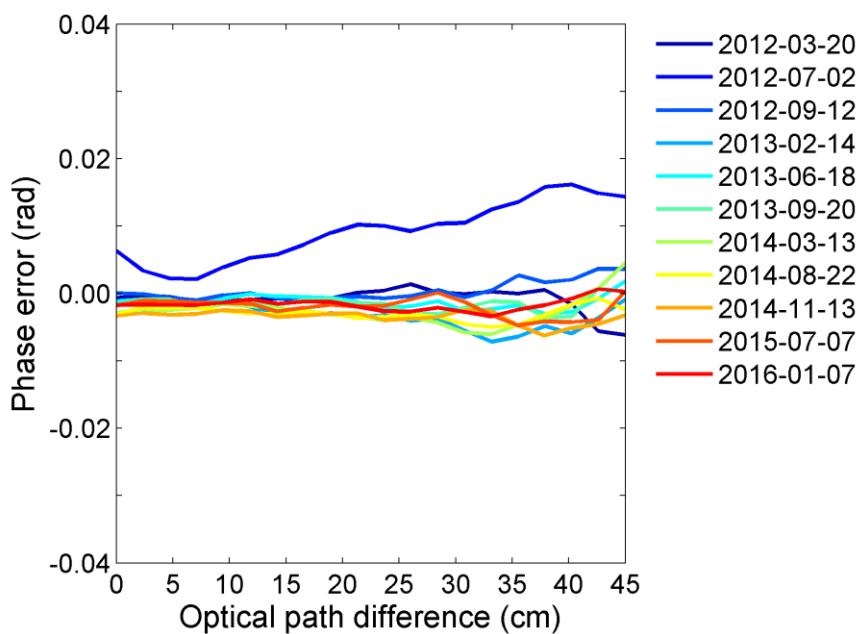
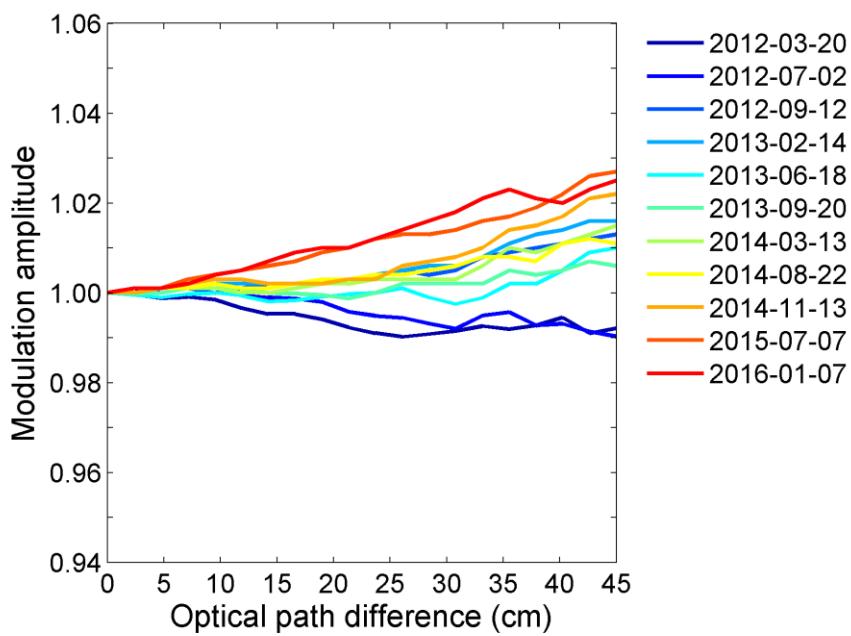
3

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 <sup>-1</sup> ) 10 <sup>-4</sup>	SPR 10 <sup>-4</sup>
up to 9.3.2010	ECL02	V01	0.4	-	4315	5	7.4	8.1
						10	13	8.2
						<b>20</b>	<b>26</b>	<b>7.7</b>
10.3.2010 to 2.3.2011	ECL04	V01	0.16	10	5960	7.5	0.42	8.0
						<b>10</b>	<b>0.75</b>	<b>8.1</b>
						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
						<b>10</b>	<b>0.14</b>	<b>8.4</b>
						20	0.56	8.4
						40	1.3	8.3

3

4

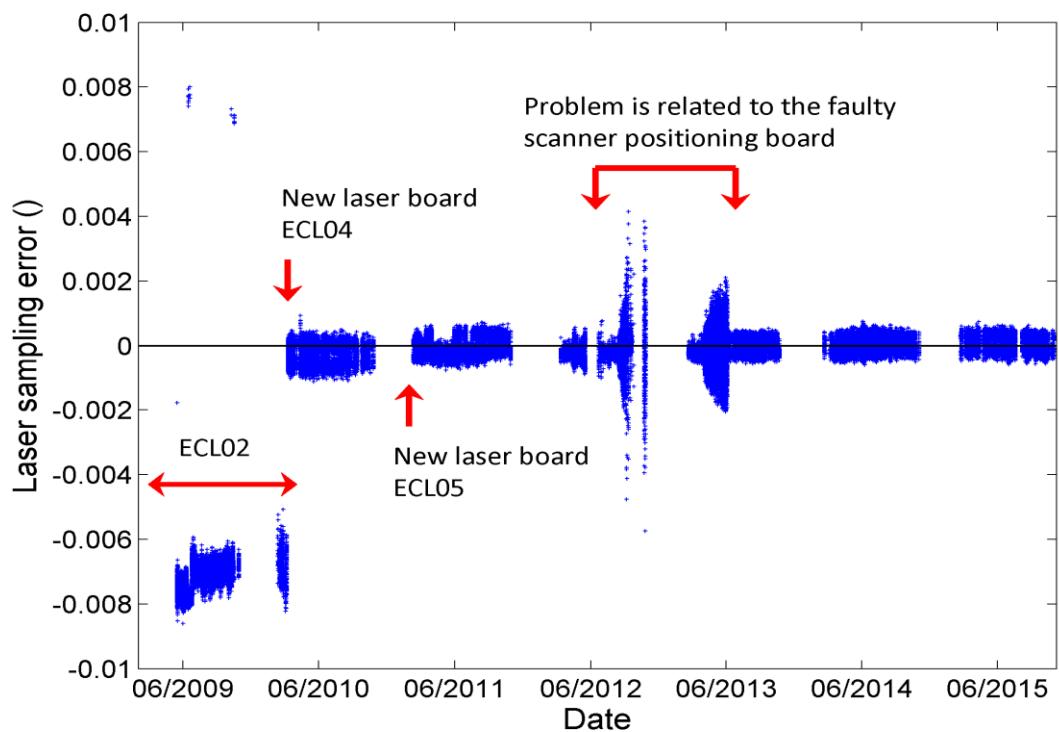


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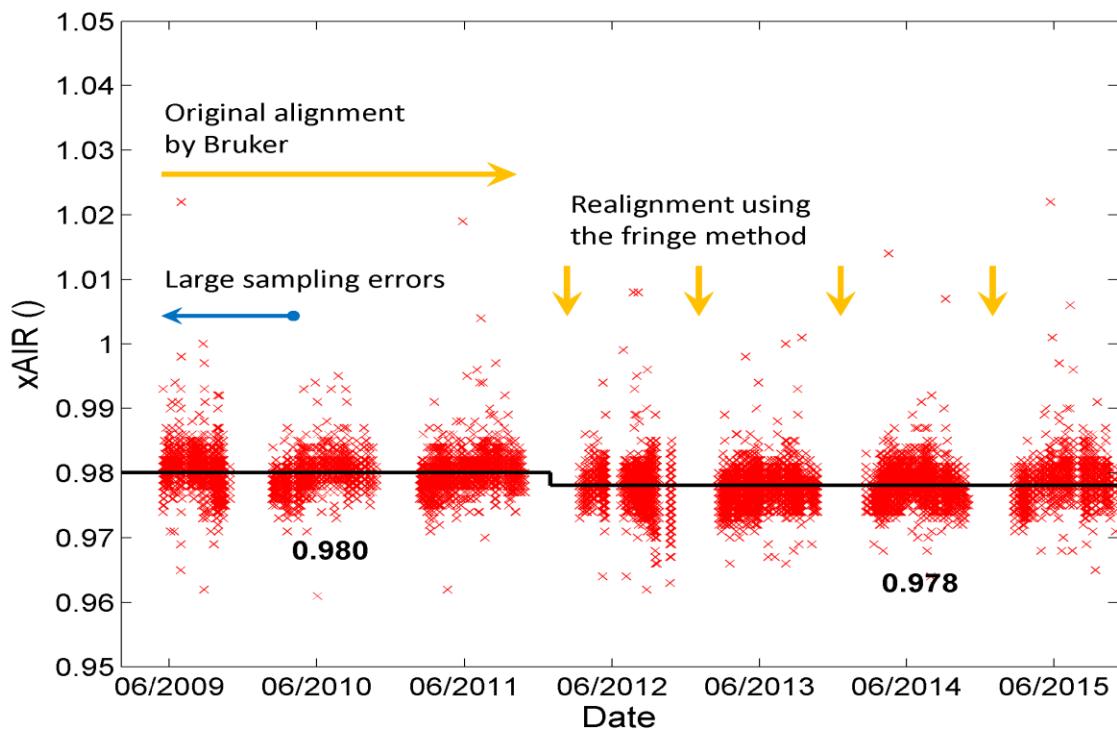
2

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

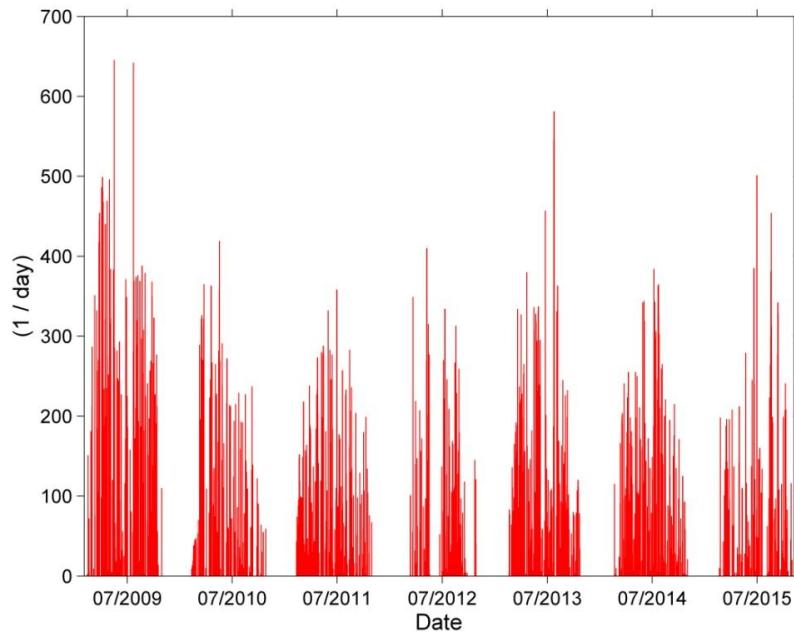
5



1  
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3 Figure 2. Laser sampling error (LSE) since 2009. LSE correction is applied during the  
4 retrieval process within GGG2014.  
5



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2  
3 Figure 3. Time series of xAIR. Average xAIR values are shown for 2009-2011 (0.980) and  
4 for 2012-2015 (0.978).  
5



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

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7

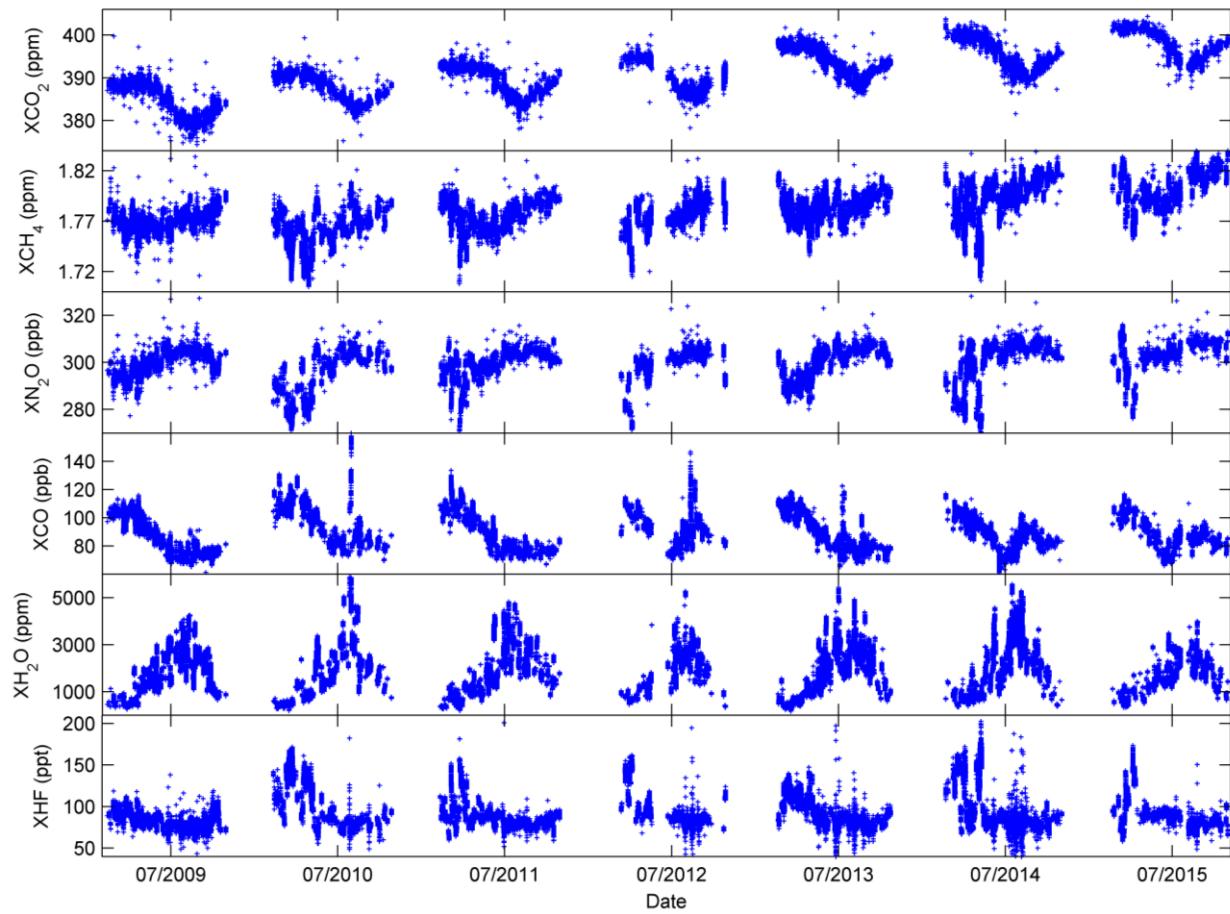
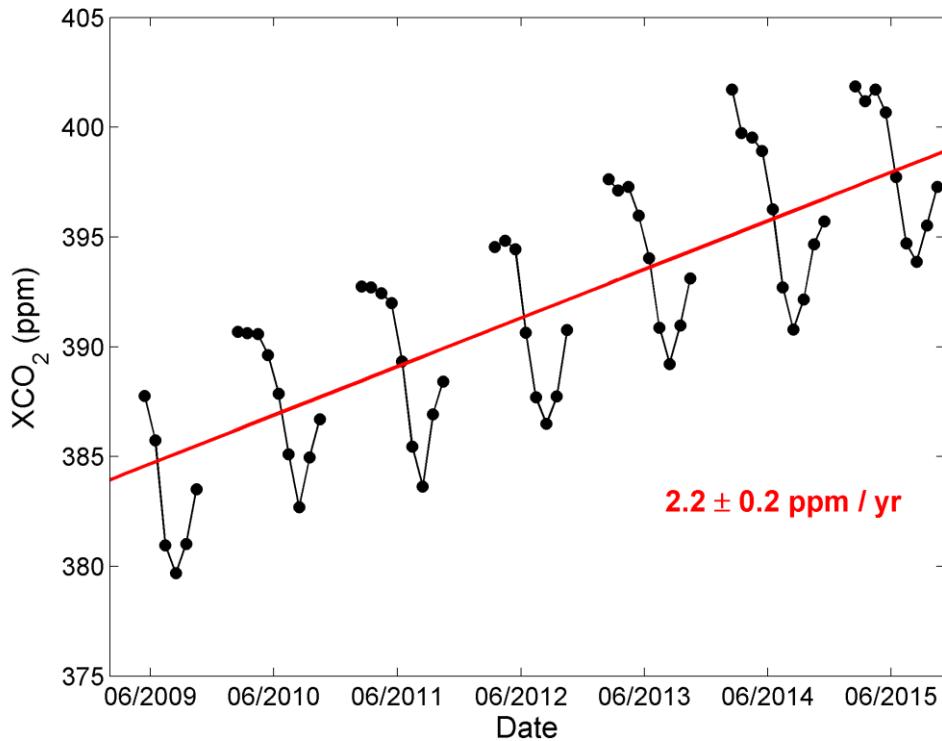


Figure 5. Time series of XCO<sub>2</sub> measurements at Sodankylä since May 2009 (upper panel). Each marker indicates a single measurement. Lower panels correspond to other gases retrieved from the same measurements.

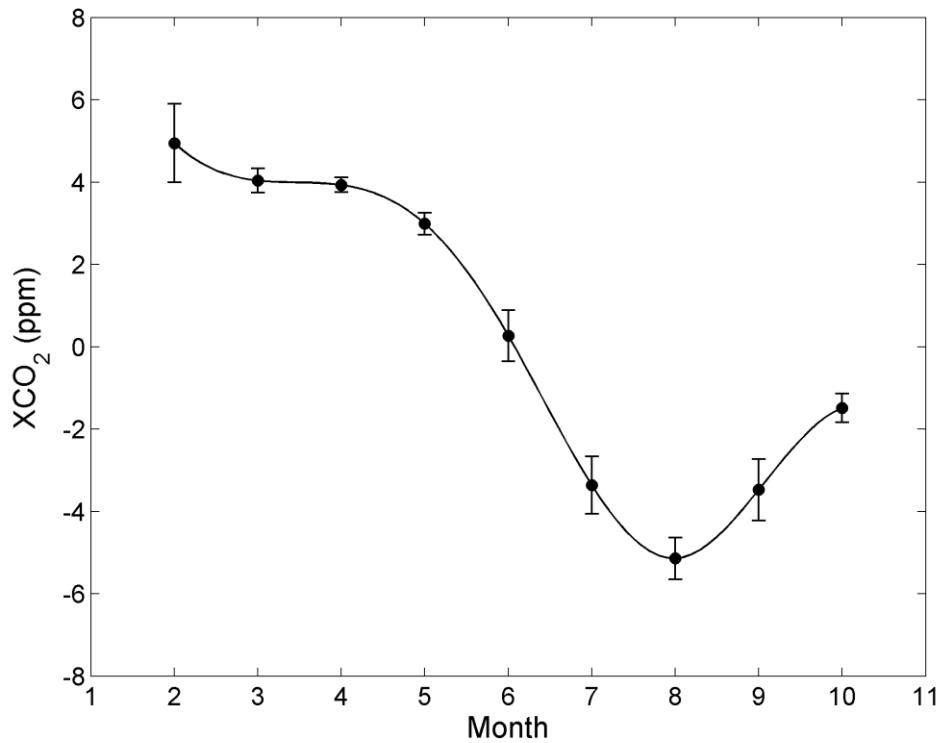
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4 Figure 6. Time series of XCO<sub>2</sub> measurements at Sodankylä since May 2009. Each marker  
 5 indicates monthly mean. A trend of 2.2+/-0.2 ppm per year has been observed during 2009-  
 6 2015.

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4 Figure 7. Average seasonal cycle of XCO<sub>2</sub> 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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