



# Using XCO<sub>2</sub> retrievals for assessing the long-term consistency of NDACC/FTIR data sets

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**Abstract.** Within the NDACC (Network for the Detection of Atmospheric Composition Change), more than 20 FTIR (Fourier-transform infrared) spectrometers, spread worldwide, provide long-term data records of many atmospheric trace gases. We present a method that uses measured and modelled XCO<sub>2</sub> for assessing the consistency of these NDACC data records. Our XCO<sub>2</sub> retrieval setup is kept simple so that it can easily be adopted for any NDACC/FTIR-like measurement made since the late 1950s. By a comparison to coincident TCCON (Total Carbon Column Observing Network) measurements, we empirically demonstrate the useful quality of this suggested NDACC XCO<sub>2</sub> product (empirically obtained scatter between TCCON and NDACC is about 4‰ for daily mean as well as monthly mean comparisons, and the bias is 25‰). Our XCO<sub>2</sub> model is a simple regression model fitted to CarbonTracker results and the Mauna Loa CO<sub>2</sub> in situ records. A comparison to TCCON data suggests an uncertainty of the model for monthly mean data of below 3‰. We apply the method to the NDACC/FTIR spectra that are used within the project MUSICA (multi-platform remote sensing of isotopologues for investigating the cycle of atmospheric water) and demonstrate that there is a good

consistency for these globally representative set of spectra measured since 1996: the scatter between the modelled and measured XCO<sub>2</sub> on a yearly time scale is only 3‰.

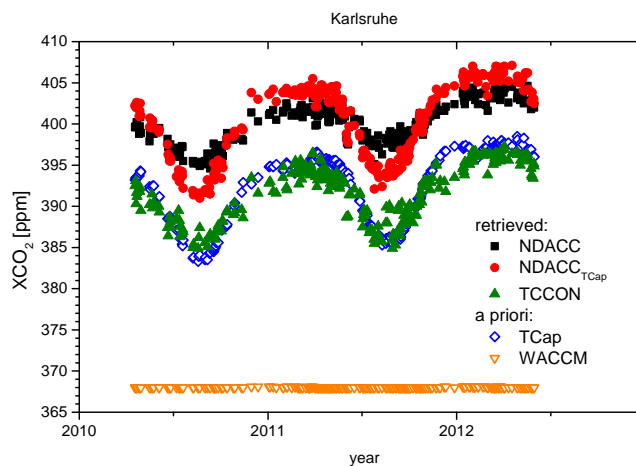
## 1 Introduction

The Network for the Detection of Atmospheric Composition Change (NDACC – formerly called Network for the Detection of Stratospheric Change, NDSC) first started measurements of atmospheric components in 1991 (Kurylo, 1991). The network is composed of more than 70 high-quality, remote-sensing research stations. Initially, the main focus was on stratospheric species and on observing the long-term change of the ozone layer, subsequently, the tropospheric composition and its link to climate change also became an important topic. Within this network, the Infrared Working Group (IRWG) operates more than 20 ground-based Fourier transform infrared (FTIR) spectrometers spread worldwide that measure the absorption of direct sunlight by atmospheric gases in the middle infrared (MIR). The strength of the NDACC is the fact that it is a network that offers numer-

ous long time series of many species and at globally distributed sites. There are many studies dealing with the long-term records of several trace gases measured within this network, e.g. ClONO<sub>2</sub>, HCl and HF (e.g. Rinsland et al., 2003; Kohlhepp et al., 2012), H<sub>2</sub>O (Schneider et al., 2012), CH<sub>4</sub> (Sussmann et al., 2012; Sepúlveda et al., 2014), N<sub>2</sub>O (Angelbratt et al., 2011a), CO and C<sub>2</sub>H<sub>6</sub> (Angelbratt et al., 2011b) and O<sub>3</sub> (Vigouroux et al., 2008). These studies would strongly benefit from a tool that is able to prove the multi-year stability of instrument precision and accuracy, later on referred to as the long-term data consistency between the different sites throughout the network. Please note that in the following the NDACC/FTIR data sets are referred to as NDACC.

In this context, it is helpful to refer to TCCON (Total Carbon Column Observing Network), which is another network of ground-based FTIR spectrometers and closely affiliated to the Infrared Working Group of NDACC. The major difference between the TCCON and NDACC is that for the former, solar spectra in the near infrared (NIR) are recorded (Wunch et al., 2011). The first TCCON measurements were obtained in 2004. The TCCON NIR spectra cover O<sub>2</sub> absorption signatures. Since atmospheric O<sub>2</sub> concentrations are very stable and well-known, TCCON O<sub>2</sub> measurements can be used for demonstrating the consistency of the TCCON measurements throughout the network. The NDACC MIR spectra do not cover O<sub>2</sub> absorption signatures and at the moment, there is no direct way to assess the long-term consistency of the NDACC FTIR measurements in analogy to TCCON. Goldman et al. (2007) attempted to use N<sub>2</sub> for this purpose. Atmospheric N<sub>2</sub> concentrations are very stable, well-known, and there are absorption signatures in the MIR between 2400 and 2450 cm<sup>-1</sup>, but due to spectroscopic issues sufficient accuracy could not be achieved.

In this paper, we propose using the total column dry-air mole fractions of CO<sub>2</sub> (XCO<sub>2</sub>) as global proxy for reviewing the long-term consistency of the MIR measurements. CO<sub>2</sub> has well isolated and easily detectable absorption signatures in the MIR spectral region. However, in contrast to O<sub>2</sub> or N<sub>2</sub>, CO<sub>2</sub> is variable with a continuing yearly increase as well as seasonal cycles with latitudinal variations, which must be accounted for in assessing the network-wide consistency. As an example, Fig. 1 shows different XCO<sub>2</sub> data sets for the Karlsruhe FTIR station. The seasonal cycle becomes clearly visible in the TCCON retrieval product (green triangles, TCCON), in the TCCON a priori (blue open diamonds, TCap), in the mid-infrared retrieval product using TCCON a priori (red dots, NDACC<sub>TCap</sub>) and in the mid-infrared retrieval product using the fixed WACCM (The Whole Atmosphere Community Climate Model, <http://waccm.acd.ucar.edu>) a priori (black squares, NDACC; the fixed WACCM a priori is shown as orange open triangles). Please note, the here called NDACC XCO<sub>2</sub> products are obtained from the retrieval setup as suggested by us for this study. They are no standard NDACC/FTIR products and thus

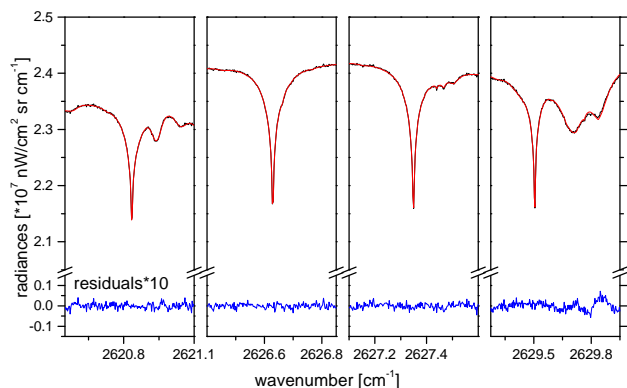


**Figure 1.** XCO<sub>2</sub> data sets for Karlsruhe, Germany (mid-latitude, Northern Hemisphere). Black: retrieved from NDACC spectra using fixed WACCM a priori, red: retrieved from NDACC spectra using TCCON a priori, green: TCCON, blue: TCCON a priori, orange: WACCM a priori.

not available via the NDACC database. The impact of the different a priori assumptions on the retrievals are discussed in detail in Sect. 2.2 and 4).

Schneider et al. (2012) showed that the de-seasonalised annual mean XCO<sub>2</sub> data obtained at 10 different NDACC/FTIR stations agree within a few per mill (when taking into account the 6 ‰ difference between the Southern and Northern Hemisphere), demonstrating that XCO<sub>2</sub> can be used as a reference for consistency for long-term measurements and for periods when many different NDACC/FTIR stations provide measurements. In this work, we further elaborate on the approach of Schneider et al. (2012). We want to check our data sets for shifts and biases caused by e.g. instrumental failure or change of instruments for the whole data sets of all used sites. Our objective is to design a method that allows an assessment of the network consistency of any NDACC/IRWG-like FTIR measurement even if it has been limited to a short campaign or to a period when the number of NDACC/FTIR stations was still relatively small (e.g. before the 2000s). For this purpose we present a simple XCO<sub>2</sub> retrieval method that can easily be adopted to any site where an NDACC-like measurement has been made since the 1950s. The measured XCO<sub>2</sub> data are then referenced to a multi-regression XCO<sub>2</sub> model that provides information on the long-term, seasonal, and latitudinal behaviour of XCO<sub>2</sub>.

In the following section, we present our simple XCO<sub>2</sub> retrieval strategy for NDACC/FTIR spectral data and briefly discuss the main differences to the more complex TCCON XCO<sub>2</sub> retrieval setup. Section 3 contains a description of our XCO<sub>2</sub> model. We demonstrate the accuracy of the model by a comparison to TCCON data. In Sect. 4, we perform an empirical validation of our NDACC XCO<sub>2</sub> data, using coincident TCCON data as a reference. In Sect. 5, the XCO<sub>2</sub> model and



**Figure 2.** The four spectral microwindows used for the ground-based FTIR retrieval. Shown is an example of a typical measurement at Karlsruhe (04 June 2010, 07:36 UT, solar elevation 38.15°). (26 October 2010, 08:44 UT, solar elevation: 18.1°). Black: measured spectrum, red: simulation and residuals (difference between measurement and simulation) multiplied by a factor of 10 (blue).

the NDACC measurements are used for demonstrating the consistency of the observations made at 10 globally representative FTIR sites.

This study uses more than 17000 individual observations made since 1996 on more than 6000 days and during different periods recorded at 10 globally distributed NDACC/FTIR sites. These are the sites that are currently contributing to the MUSICA project (multi-platform remote sensing of isotopologues for investigating the cycle of atmospheric water, Schneider et al., 2012). The paper concludes with a summary and an outlook.

## 2 The XCO<sub>2</sub> NDACC retrieval

### 2.1 The CO<sub>2</sub> retrieval setup

The ground-based FTIR systems measure solar absorption spectra using high-resolution Fourier transform spectrometers. For our analysis we apply the retrieval code PROFIT and the included radiative transfer code PROFFWD (Hase et al., 2004). PROFIT has been used for many years in the ground-based FTIR community for evaluating high-resolution solar absorption spectra. Details about the retrieval principles are described by e.g. Schneider et al. (2012) or Sepúlveda et al. (2012).

The spectral microwindows that are used for this study are shown in Fig. 2 and Table 1 (Kohlhepp, 2007). In addition to CO<sub>2</sub> we have considered spectroscopic signatures of the interfering species H<sub>2</sub>O and CH<sub>4</sub>. The spectroscopic line parameters for CO<sub>2</sub> and CH<sub>4</sub> have been taken from the HITRAN (High-Resolution Transmission Molecular Absorption) 2008 database (Rothman et al., 2009), while for H<sub>2</sub>O we applied the HITRAN 2009 update ([www.cfa.harvard.edu/hitran/](http://www.cfa.harvard.edu/hitran/)).

**Table 1.** Spectral microwindows chosen for the NDACC CO<sub>2</sub> retrieval shown in this study.

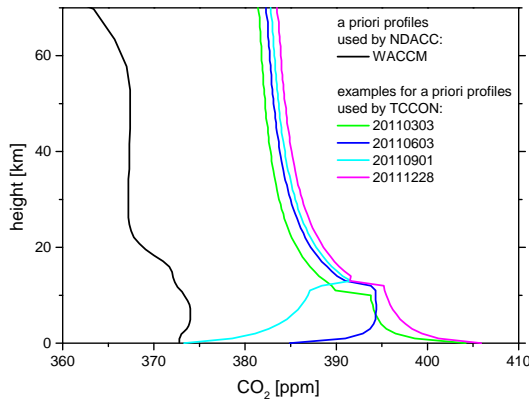
	Spectral microwindows (cm <sup>-1</sup> )
MW1	2620.550–2621.100
MW2	2626.400–2626.850
MW3	2627.100–2627.600
MW4	2629.275–2629.950

### 2.2 Profile scaling and a priori information

An important aspect of our study is that we use a rather simple retrieval setup (fixed a priori and simple scaling retrieval). This assures that the method can be correctly and consistently applied for different sites and time periods, without any risk of inconsistency due to a priori and constraints. The spectral windows, as depicted in Fig. 2, contain some weak H<sub>2</sub>O, HDO and CH<sub>4</sub> lines. In order to minimise the spectral interferences due to the highly variable atmospheric amounts of H<sub>2</sub>O and HDO, we investigated a two-step retrieval strategy. First, the H<sub>2</sub>O profile is determined by the MUSICA H<sub>2</sub>O retrieval (Schneider et al., 2010, 2012, 2015). As a second step, CO<sub>2</sub> is retrieved by simultaneous scaling with its interfering species CH<sub>4</sub> and H<sub>2</sub>O. Here, the retrieved daily mean H<sub>2</sub>O profile as a result of the first step is used as a priori information. However, we found that the H<sub>2</sub>O and HDO absorptions are rather weak so that not applying the two-step strategy does not significantly affect the CO<sub>2</sub> results. For CO<sub>2</sub> and CH<sub>4</sub>, we apply the climatological entries from WACCM version 6, provided by NCAR (National Center for Atmospheric Research, J. Hannigan, private communication, 2009). The WACCM a priori profile of CO<sub>2</sub> for Karlsruhe is the black line in Fig. 3. We use this single WACCM a priori profile for all the NDACC CO<sub>2</sub> retrievals made at Karlsruhe (Fig. 1). The WACCM simulations can vary between sites, but for each site a temporally constant a priori is applied. This is a main difference with respect to the TCCON retrieval setup, which is much more complex in this context. The CO<sub>2</sub> a priori information used by the TCCON retrieval for the actual TCCON data set varies from day to day, which has to be properly considered for comparisons with other data sets.

For the TCCON retrievals the CO<sub>2</sub> a priori information varies from day to day, which has to be properly considered if one wants to setup a TCCON-like XCO<sub>2</sub> retrieval. The coloured lines in Fig. 3 represent the TCCON a priori profiles for Karlsruhe for 4 days in 2011 during different seasons that are used to calculate the TCCON data set (Sect. 3.2.1).

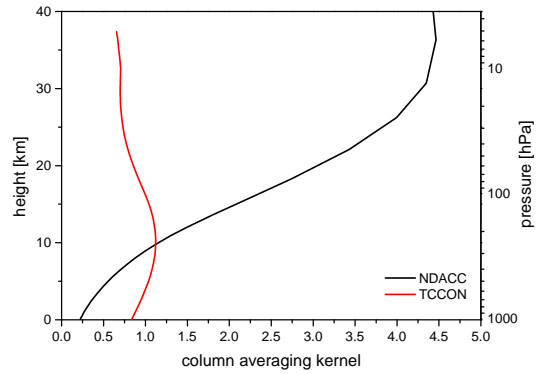
The a priori assumptions affect the retrieval results. For sites where NDACC and TCCON measurements are made simultaneously, we made two different retrievals with the NDACC MIR spectra. First we applied our simple retrieval recipe (fixed WACCM a priori), and then we calculated the results when using the TCCON strategy (daily varying a pri-



**Figure 3.** A priori profiles for Karlsruhe: WACCM v6 (used as a priori for NDACC retrieval) and some examples used as a priori for TCCON retrieval.

ori). Figure 1 shows the different retrieval results for Karlsruhe. The NDACC XCO<sub>2</sub> values obtained with the fixed WACCM a priori and the varying TCCON a priori are depicted as the orange and red dots, respectively. In addition, the figure shows the TCCON XCO<sub>2</sub> product (green dots). The a priori XCO<sub>2</sub> assumptions are plotted as blue and orange dots (for the fixed WACCM and the varying TCCON assumptions, respectively). The influence of the a priori information on the series and especially on the seasonal cycle will be further discussed in Sect. 4. Typical column-averaging kernels for NDACC and TCCON which represent the height-dependent sensitivity of the retrieved total column to perturbations of the partial columns at the various atmospheric levels are shown in Fig. 4. A value of 4.5 at 30 km means, that an input disturbance at these altitude levels would be significantly overestimated, which is a consequence of the lower values at heights between ground and 5 km. However note that the variability in the stratosphere is very small. An ideal column-averaging kernel would be unitary (Connor et al., 2008). Compared to TCCON, the NDACC spectra have less sensitivity in the lower troposphere, which means that small variations in that region won't be captured in detail. However, in the following we will show that this is sufficient for our study as we are interested in variabilities on timescales longer than a month. On these timescales, the model we use (Sect. 3) is valid, so the difference between NDACC XCO<sub>2</sub> and the model can be used as proxy for the stability of the NDACC data.

NCEP (National Centers for Environmental Prediction) analysis data at 12 UT are used for daily temperature and pressure profiles for all sites (e.g. Lait, 2015).



**Figure 4.** Column-averaging kernel for Karlsruhe (NDACC; 04 June 2010, 07:36 UT, solar elevation 38.15°) and Lamont (TCCON) for the same solar elevation.

**Table 2.** Uncertainty sources used for our error estimation. The second column gives the assumed uncertainty value and the third column the assumed partitioning between statistical and systematic sources.

Error source	Uncertainty	Stat./Syst.
Baseline (channelling/offset)	0.02 %/0.1 %	50/50
Instrumental line shape (Mod. eff./phase error)	1 %/0.01 rad	50/50
Temperature profile	2–5 K	70/30
Line of sight	0.1°	90/10
Solar lines (Intensity/ $\nu$ -scale)	1 %/ $10^{-6}$	80/20
Spectroscopic parameters ( $S/\gamma$ )	2/5 %	0/100

### 2.3 Error estimation

The assumptions we made for our error calculations are listed in Table 2. Apart from uncertainties in the spectroscopic parameters (line strength  $S$  and pressure broadening  $\gamma$ ), which are purely systematic, we assume a systematic and a statistical contribution for each uncertainty. An exception is the error due to measurement noise, which we assume to be purely random.

The errors, estimated with the error calculation implemented in PROFFIT for a typical measurement at Karlsruhe station (04 June 2010, 07:36 UT, solar elevation 38.15°), are listed in Table 3. The systematic error is clearly dominated by spectroscopy, mainly due to the pressure broadening error. The leading random error source is the measurement noise followed by the baseline uncertainty.

### 2.4 Calculation of XCO<sub>2</sub>

NDACC XCO<sub>2</sub> is calculated by dividing the CO<sub>2</sub> total column by the dry pressure column (DPC). The DPC is obtained by converting the ground pressure to column air concentra-

**Table 3.** Statistical and systematic errors in the Karlsruhe total CO<sub>2</sub>-column due to the assumed uncertainty sources of Table 2. The total error represents the root-sum-squares of all errors.

Error	Statistical [%]	Systematic [%]
Measurement noise	0.20	
Baseline	0.17	0.17
Instrumental line shape	0.02	0.02
Temperature	0.10	0.07
Line of sight	0.04	0.01
Solar lines	0.02	0.01
Spectroscopy		4.22
TOTAL	0.33	4.23

tion (e.g. Deutscher et al., 2010):

$$\text{DPC} = \frac{P_s}{m_{\text{dryair}} \cdot g(\varphi)} - \frac{m_{\text{H}_2\text{O}}}{m_{\text{dryair}}} \cdot \text{H}_2\text{O}_{\text{col}} \quad (1)$$

where  $P_s$  is the surface pressure in Pascals,  $m_{\text{dryair}}$  the molecular mass of the dry air ( $\sim 28.96 \times 10^{-3} \text{ N}_A \text{ kg molecule}^{-1}$ ),  $m_{\text{H}_2\text{O}}$  the molecular mass of the water vapour ( $\sim 18 \times 10^{-3} \text{ N}_A \text{ molecule}^{-1}$ ),  $\text{H}_2\text{O}_{\text{col}}$  the water vapour total column amount (in molecules  $\text{m}^{-2}$ ,  $\text{N}_A$  Avogadro's constant ( $\sim 6.022 \times 10^{23} \text{ molecules mol}^{-1}$ ) and  $g(\varphi)$  the latitude-dependent surface acceleration due to gravity.  $\text{H}_2\text{O}_{\text{col}}$  is a result of the MUSICA retrieval (Schneider et al., 2010, 2012) and surface pressure is taken from NCEP.

This method is not expected to be as precise as the TCCON method which uses the measured O<sub>2</sub> column as reference when calculating the dry pressure column. However, it has the advantage that it can be easily applied to all historic measurements made in the 2600–3000  $\text{cm}^{-1}$  spectral region (both H<sub>2</sub>O and CO<sub>2</sub> can be retrieved in the same filter region so there is no need for additional coinciding measurements).

### 3 The XCO<sub>2</sub> model

#### 3.1 Description of the model

As reference for the XCO<sub>2</sub> measurements we use an empirical XCO<sub>2</sub> model. The empirical model formula includes a polynomial fit with an empirical lookup adjustment to the Mauna Loa series (<http://co2now.org/Current-CO2/CO2-Now/scripps-co2-data-mauna-loa-observatory.html>) (Keeling et al., 2001, 2005), considering global trace gas distribution given by CarbonTracker (<http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/>) (Peters et al., 2007). The formula is a modification of that used in Reuter et al. (2012), suggested by F. Hase (private communication, 2012), allowing the calculation of reasonable values with an improved latitude-dependency also outside the fit period.

The only inputs required for the model in addition to the Mauna Loa time series and CarbonTracker are the time, lat-

itude and typical surface pressure of the measurement site. The XCO<sub>2</sub> is modelled via

$$y(t, l_r, P_{\text{stat}}) = c_0 + d_{\text{NDACC}} \cdot d_h \cdot \text{ML}_a (a_{2\pi} \sin(2\pi t + \phi_{2\pi}) + a_{4\pi} \sin(4.0\pi t + \phi_{4\pi})), \quad (2)$$

where  $t$  is the decimal year,  $l_r$  the latitude in rad and  $P_{\text{stat}}$  the typical pressure at the measurement site.  $c_0$  contains the time- and latitude-dependent CO<sub>2</sub>-increase:

$$c_0 = (\text{ML}_{\text{smooth}} + \text{ML}_{\text{corr}}) (e_1 + e_2),$$

where  $\text{ML}_{\text{smooth}}$  describes the inter-annual increase of CO<sub>2</sub>:

$$\text{ML}_{\text{smooth}} = 316.5 + 0.8407 t_d + 0.012 t_d^2,$$

with  $t_d = t - 1960$ .  $\text{ML}_{\text{corr}}$  are yearly correction factors that consider the year-to-year variations of the Mauna Loa series, compared to a polynomial fit. All  $\text{ML}_{\text{corr}}$  values are listed in Appendix C (Table C1). The coefficients  $e_1$  and  $e_2$  construct latitudinal gradients on the predicted long-term CO<sub>2</sub> concentration:

$$e_1 = a_1 + \frac{a_2}{\exp^{2.5(l_r+0.2)} + 1.0}$$

$$e_2 = \frac{a_3}{\exp^{-6.0(l_r-0.9)} + 1.0}$$

with  $a_1 = 1.0018$ ,  $a_2 = -0.0106576$  and  $a_3 = -2.132 \exp^{-3}$ . The amplitude of the Mauna Loa series is described by

$$\text{ML}_a = 0.5 (3.0 + 0.006 (t - 1959.0)).$$

As in Reuter et al. (2012), the seasonal cycle has a 12- and a 6-month period with a latitudinal-dependent phase:

$$a_{2\pi} = 0.8 \left( 0.2 + \frac{4.0}{\exp^{-3.5(l_r-0.5)} + 1.0} \right)$$

$$a_{4\pi} = 0.5 \left( 0.05 + \frac{1.7}{\exp^{-3.5(l_r-0.5)} + 1.0} \right)$$

$$\phi_{2\pi} = -2.75 + \frac{3.3}{\exp^{-2.8(l_r+0.2)} + 1.0}$$

$$\phi_{4\pi} = 1.45 + 1.9 \sin(1.4 l_r).$$

To consider the influence of the measurement height on the seasonal cycle, we implemented the damping factor  $d_h$ :

$$d_h = \frac{(P_{\text{stat}} - P_{\text{TP}})}{(P_{\text{grnd}} - P_{\text{TP}})}, \quad (3)$$

where  $P_{\text{TP}}$  is the typical tropopause pressure for the site latitude and  $P_{\text{grnd}} = 1013.25 \text{ hPa}$ .

For approximating the actual atmospheric XCO<sub>2</sub> values,  $d_{\text{NDACC}}$  is set to unity. For reproducing NDACC-type CO<sub>2</sub> observations, it is a smaller value than unity (accounting for the non-ideal column sensitivity of the retrieval; for details see Sect. 4.2).



**Table 4.** Overview of collaborating ground-based NDACC/TCCON FTIR stations.

Site	Country	Latitude	Longitude	Height [m a.s.l.]	Institution
Eureka (NDACC + TCCON)	Canada	80.1° N	86.4° W	610	U. Toronto
Ålesund (NDACC + TCCON)	Norway	78.9° N	11.9° E	21	U. Bremen/AWI
Kiruna (NDACC)	Sweden	67.8° N	20.4° E	419	KIT IMK-ASF/IRF
Sodankylä (TCCON)	Finland	67.4° N	26.6° E	188	FMI ARC
Bremen (NDACC + TCCON)	Germany	53.1° N	8.9° E	27	U. Bremen
Karlsruhe (TCCON + MIR)	Germany	49.1° N	8.4° E	110	KIT IMK-ASF
Jungfraujoch (NDACC)	Switzerland	46.6° N	8.0° E	3580	U. Liège
Izaña (NDACC + TCCON)	Spain	28.3° N	16.5° W	2367	KIT IMK-ASF/AEMET
Wollongong (NDACC + TCCON)	Australia	34.5° S	150.9° E	30	U. Wollongong
Lauder (NDACC + TCCON)	New Zealand	45.1° S	169.7° E	370	NIWA
Arrival Heights (NDACC)	Antarctica	77.8° S	166.7° E	250	NIWA

Due to the fact that the model does not include meteorological fields, the calculated values can only be valid on a monthly time scale and not on a synoptic or daily time scale. To account for that, we only compare monthly mean data, which are calculated from daily means and we require that the standard error of the so calculated monthly mean is smaller than 5%.

### 3.2 Empirical uncertainty assessment of the model

As in Reuter et al. (2012), we use the TCCON data set for validating our model. In the following, we briefly introduce TCCON and discuss the quality of the TCCON XCO<sub>2</sub> data, after that we compare our model calculation to the TCCON reference.

#### 3.2.1 The TCCON XCO<sub>2</sub> data set

TCCON is a network of ground-based Fourier transform spectrometers that record direct solar spectra in the NIR. It was founded in 2004 and operates around 20 spectrometers spread worldwide. From these spectra, accurate and precise column-averaged abundances of atmospheric constituents including CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, HF, CO, H<sub>2</sub>O and HDO, are retrieved.

To retrieve trace gas columns from the measured spectra, the software package GGG2012, developed by G. Toon (JPL), is used (Wunch et al., 2011, 2012). For CO<sub>2</sub> a profile scaling retrieval approach is applied. As interfering species, HDO, H<sub>2</sub>O and CH<sub>4</sub> are considered. There are two selected windows: the central wave numbers are 6220.0 and 6339.5 cm<sup>-1</sup> with spectral widths of 80 and 85 cm<sup>-1</sup>, respectively. The spectroscopic data of Toth et al. (2008) and Rothman et al. (2009) with empirical extensions of G. Toon (personal communication, 2014) are used.

TCCON uses time-dependent CO<sub>2</sub> a priori profiles. Up to 10 km, the a priori profile is the result of an empirical model, based on fits to GLOBALVIEW data and independent vertical profiles (e.g. AirCore, aircraft overflights). In the strato-

sphere, an age-dependent profile is assumed. Examples of TCCON a priori profiles for different seasons are shown in Fig. 3. Already this empirical a priori model provides a good estimator for the actual atmospheric XCO<sub>2</sub> and the difference between the a priori model and the TCCON result is often smaller than 1% (typical scatter between blue and green symbols in Fig. 1). The objective of TCCON is to significantly improve the estimations of the model and get an accuracy for XCO<sub>2</sub> of better than 2%. Such high and network-wide precision is considered mandatory for using XCO<sub>2</sub> for carbon cycle research (Olsen and Randerson, 2004).

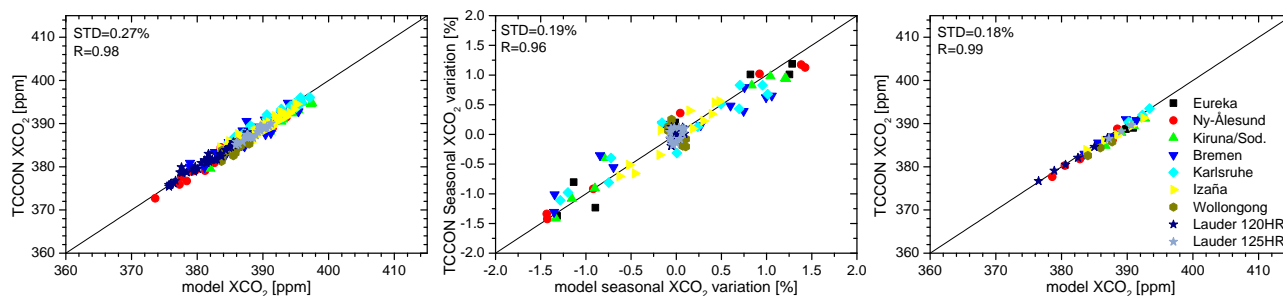
TCCON data products are column-averaged dry-air mole fractions, which for e.g. XCO<sub>2</sub> are calculated as (Wunch et al., 2011):

$$\text{XCO}_2 = 0.2095 \frac{\text{CO}_2^{\text{col}}}{\text{O}_2^{\text{col}}} . \quad (4)$$

Taking the ratio to the co-observed O<sub>2</sub> column has the advantage that correlated errors of CO<sub>2</sub> and O<sub>2</sub> are reduced. The time-dependent TCCON a priori assumptions cause a pronounced annual cycle and inter-annual trend in the TCCON XCO<sub>2</sub> a priori data (blue dots in Fig. 1).

There have been several calibration campaigns (Washenfelder et al., 2006; Deutscher et al., 2010; Wunch et al., 2010, 2012; Messerschmidt et al., 2011) that all consistently yield a calibration factor of  $0.989 \pm 0.001$  for XCO<sub>2</sub>. The data we use in this study have all been corrected by this calibration factor.

TCCON data sets (GGG2012) have been downloaded from the TCCON database (<http://tcon.ipac.caltech.edu/>). Due to the fact that some of these measurements have been recorded with faulty laser sampling boards (Messerschmidt et al., 2010), they were biased relative to the data recorded after the board exchange. Dohe et al. (2013) have developed a re-sampling algorithm that has been either already applied to most of the affected data before the upload or the effect was negligible (TCCON, 2013). Of the data sets used, only the time series of Bremen and Wollongong had to be manu-



**Figure 5.** Correlation of TCCON vs. XCO<sub>2</sub> model for the time period 2005–2012. Left: monthly means, middle: detrended seasonal cycles, right: de-seasonalised yearly means. Added is the one-to-one correlation (black line). At Lauder, two TCCON instruments have been used (Bruker 120HR/125HR).

ally corrected before our comparisons. In the case of Bremen, all measurements made before 18 June 2009 needed a correction of  $-1.2$  ppm; the Wollongong data measured until 22 July 2011 have been shifted by  $-1.0 \pm 1$  ppm. A detailed description of this error can be found on the TCCON web site (TCCON, 2013) as well as in Dohe et al. (2013) and Messerschmidt et al. (2010).

### 3.2.2 Comparison of modelled XCO<sub>2</sub> and TCCON XCO<sub>2</sub>

The TCCON sites have been chosen to fit to our actual set of NDACC sites (Table 4). At Eureka, Ny-Ålesund, Bremen, Karlsruhe, Izaña, Wollongong and Lauder NDACC and TCCON measurements are performed at the same site. At Kiruna, Jungfraujoch and Arrival Heights there are no TCCON measurements. However, Kiruna is located close to the TCCON station of Sodankylä ( $67.3668^\circ$  N,  $26.631^\circ$  E,  $0.188$  m a.s.l.), which is 260 km east and around 40 km south of Kiruna. Therefore, the Sodankylä TCCON data is paired with the Kiruna NDACC and the modelled XCO<sub>2</sub> data.

Figure 5 shows the correlation of TCCON versus the modelled XCO<sub>2</sub> data set for the time period 2005–2012 for all the TCCON sites we work with in this study. The left graph shows the correlation for monthly mean data, which is the time scale that can be reproduced by the model (please recall that it cannot capture synoptic time scale variations). The correlation coefficient is  $R = 0.98$  and the scatter (SD of the difference between model and measurement) is  $2.7\%$ . We investigate this agreement in more detail by looking at different time scales: the seasonal cycle (or intra-annual time scale) and the long-term evolution (or inter-annual time scale).

The middle graph of Fig. 5 compares the measured and modelled detrended seasonal cycles calculated for each site. It is expressed in percentage values because it is the seasonal variation with respect to the de-seasonalised data. For its calculation we first remove the inter-annual trend and then calculate the mean for each month of the year (the inter-annual trend is removed by fitting a Fourier series according to e.g.

Gardiner et al., 2008). We observe that the model captures well the variation on this seasonal time scale ( $R = 0.96$  and a scatter of  $1.9\%$ ). Detailed documentation of the seasonal cycles for the different sites can be found in Appendix A (Fig. A1).

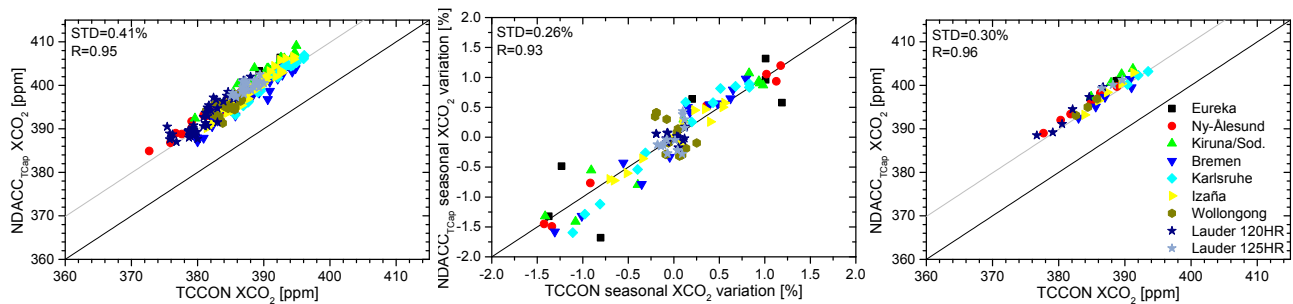
The correlation of the de-seasonalised yearly means is plotted in the right graph (it is the yearly mean as calculated from the data after removing the seasonal cycle). On this inter-annual time scale the agreement is very good ( $R = 0.99$  and a scatter of  $1.8\%$ ).

Overall, the model and measurements agree very well. According to the scatter between the model and the TCCON data, the model is able to predict the XCO<sub>2</sub> amounts on a monthly time scale with a precision of better than  $3\%$  and on a yearly time scale with a precision of better than  $2\%$ .

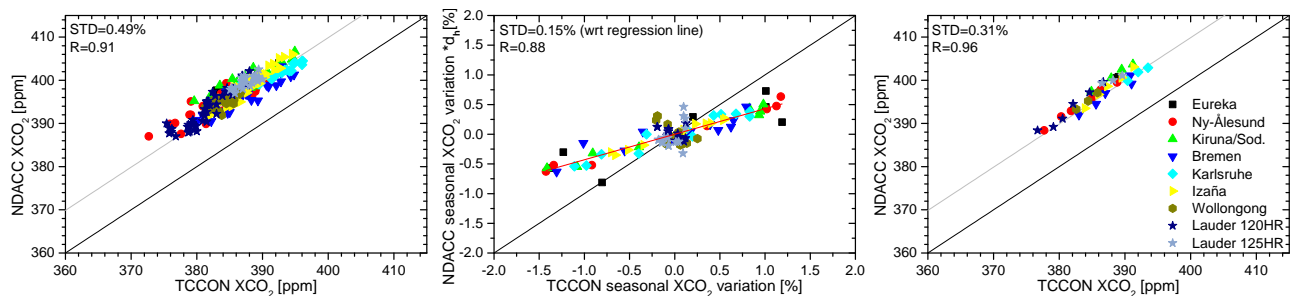
## 4 Empirical validation of the NDACC XCO<sub>2</sub> data

In this section, we want to check the quality of the NDACC XCO<sub>2</sub> data. As for the validation of the model (see previous Section), we use the TCCON XCO<sub>2</sub> data set as reference to check the quality and the influence of the retrieval strategy on the data set. When comparing TCCON and our XCO<sub>2</sub> product, we have to be aware that the retrieval strategies are different: while for our NDACC retrieval we use a fixed a priori for each site, for TCCON the a priori is changing from day-to-day. The empirical validation is made for the eight sites of our study, where TCCON and NDACC measurements are made at the same site (or nearby, as in Kiruna/Sodankylä) and on the same day (we use daily mean data as the basis for the comparison).

In the following, we present two types of comparison. First, we generate an XCO<sub>2</sub> product from NDACC measurements that uses the same varying a priori information as the TCCON retrieval, hereinafter called NDACC<sub>TCap</sub>. Both the TCCON and the NDACC<sub>TCap</sub> XCO<sub>2</sub> data sets are influenced by the same a priori information. This means that differences between these two data sets are rather directly linked to the different measurements (different spectral resolution



**Figure 6.** As Fig. 5 but for NDACC XCO<sub>2</sub> applying the varying TCCON a priori (NDACC<sub>TCap</sub>) vs. TCCON. The black line represents the one-to-one correlation, the grey line is the correlation shifted by 2.5 %.



**Figure 7.** As Fig. 5 but for NDACC XCO<sub>2</sub> applying the fixed WACCM a priori. Left: monthly means, middle: detrended seasonal cycles with regression line (red), given standard deviation with respect to the regression line, right: de-seasonalised yearly means. The black line represents the one-to-one correlation, the grey line is the correlation shifted by 2.5 %.

and spectral region). Second, we compare the XCO<sub>2</sub> NDACC product (obtained by the fixed a priori) with the TCCON product (varying a priori). Differences in these two data sets are due to the different measurements and the different a priori assumptions. However, since the TCCON product is of a well known and high absolute quality, this second comparison exercise can reveal the actual capability of the XCO<sub>2</sub> NDACC retrieval when using a fixed a priori (recall that the advantage of this recipe is that it can be easily adopted for any site and any time period).

#### 4.1 Comparison of NDACC and TCCON XCO<sub>2</sub> when using the same varying a priori

Figure 6 shows the comparison between the NDACC<sub>TCap</sub> and the TCCON XCO<sub>2</sub> data sets. The panels from the left to the right are analogous to Fig. 5 for the different time scales: left panel for monthly mean data, central panel for detrended intra-annual monthly mean variations and right panel for de-seasonalised yearly means.

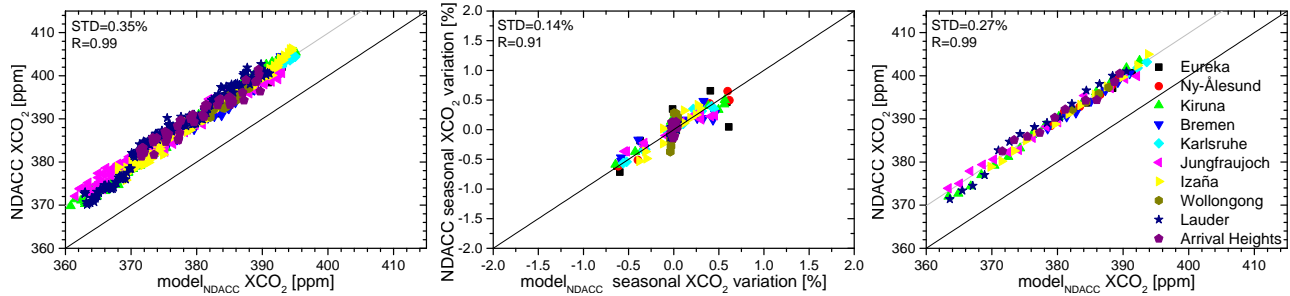
We observe a good correlation between the two data sets. The scatter is about 4 ‰ on a monthly time scale and 3 ‰ on a yearly time scale. However, there is a significant systematic difference. The NDACC XCO<sub>2</sub> values are by 25 ‰ larger than the TCCON XCO<sub>2</sub> values. There are two reasons that might explain the systematic difference. First, our NDACC XCO<sub>2</sub> product is calculated using DPC (Eq. 1) whereas for

TCCON O<sub>2</sub> is used (Eq. 4). However, the O<sub>2</sub> columns are known to be 2 % too high (Wunch et al., 2010) and applying the O<sub>2</sub> columns instead of the DPC values for the calculation of the NDACC XCO<sub>2</sub> data would yield about 2 % smaller values and thus a much smaller difference with respect to the TCCON XCO<sub>2</sub> data. Note that the TCCON calibration is made for XCO<sub>2</sub> not for CO<sub>2</sub>. Second, NDACC and TCCON measure in different spectral regions, thus part of the differences are probably caused by inconsistencies between the spectroscopic data (Sects. 2.1 and 3.2.1).

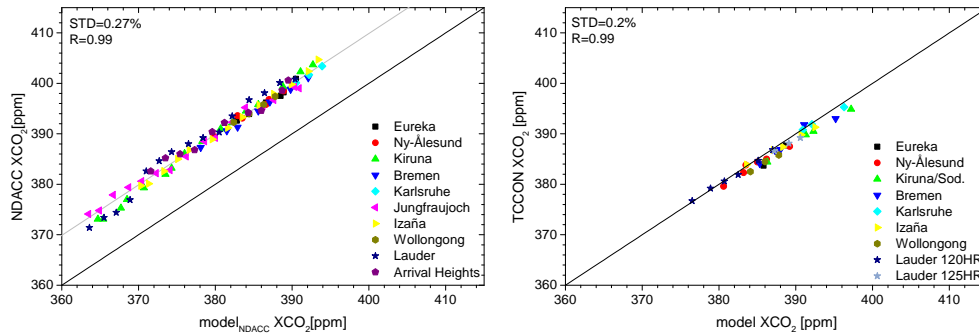
The comparison of the seasonal variations (central panel) also reveals good agreement. When using the same a priori as TCCON, the NDACC measurements can reproduce the TCCON seasonal variation within 2.6 ‰ (scatter between the two data sets). A detailed overview on the seasonal cycles for the different sites is given in the Appendix A (Fig. A1).

The main interest of this study are monthly or longer time scales, which are decisive for the reliability of trend analyses. These time scales can be well captured by the model (Sect. 3). However, often TCCON and NDACC measurements are made on the same day, and we can compare the measurements on a daily time scale. Although it is not of direct interest for our long-term study, the day-to-day NDACC vs. TCCON comparison can serve as a good measure for the quality of the NDACC XCO<sub>2</sub> product. We found that the day-to-day scatter between the NDACC and TCCON data





**Figure 8.** As Fig. 5 but for NDACC XCO<sub>2</sub> applying the WACCM fixed a priori and model<sub>NDACC</sub> and for the time period 1996–2012.



**Figure 9.** Correlations of the yearly means. Left: NDACC vs. model NDACC (1996–2012), right: TCCON vs. model (2005–2012).

sets is generally within 4 ‰ (Appendix B). This good agreement nicely documents that the CO<sub>2</sub> signatures recorded by the NDACC spectra can serve as a reliable quality proxy for the NDACC data sets. For some sites the agreement with TCCON is even within 3 ‰. In addition, Fig. B1 shows that the NDACC XCO<sub>2</sub> data can reveal the deficits of the simple XCO<sub>2</sub> a priori model in a similar manner as the TCCON XCO<sub>2</sub> data. This suggests that the NDACC XCO<sub>2</sub> data might even be useful for carbon cycle research. For this purpose a precision for XCO<sub>2</sub> of at least 2 ‰ is required (Olsen and Randerson, 2004). This high precision can be achieved by the TCCON measurements and the results of Appendix B suggest that for some stations an NDACC product can achieve a similar high precision. A further investigation of a possible extension of the TCCON XCO<sub>2</sub> time series by NDACC XCO<sub>2</sub> data is beyond the scope of our paper and currently under investigation by M. Buschmann (University of Bremen, private communication, 2014).

#### 4.2 Comparison of NDACC (fixed a priori) and TCCON XCO<sub>2</sub> (varying a priori)

Figure 7 shows the same as Fig. 5, but for NDACC XCO<sub>2</sub> obtained when using a fixed a priori. For the comparison of de-seasonalised yearly means (right panel), we observe a very good correlation ( $R = 0.96$ ), a rather small scatter of 3.1 ‰ and a systematic difference of 25 ‰. This is almost identical to what we observe in Fig. 6 for the comparison

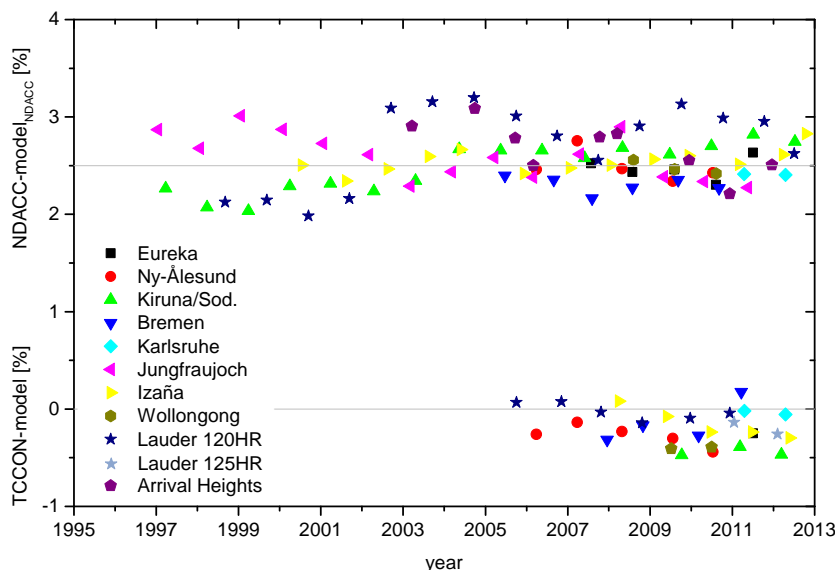
between the NDACC<sub>TCap</sub> and the TCCON XCO<sub>2</sub> data sets. The NDACC retrieval that uses a fixed a priori is sufficient to retrieve the long-term evolution of XCO<sub>2</sub>. It is not necessary to use an a priori that simulates this long-term behaviour. For the monthly mean data (left panel, Fig. 7), the correlation is poorer than observed in the left panel of Fig. 6. As explained in the following, this poorer agreement is mainly due to the failure of the NDACC XCO<sub>2</sub> data to capture the full amplitude of the seasonal cycle.

On seasonal time series, variations in the CO<sub>2</sub> profile occur mostly in the lower to middle troposphere, meaning that the variation occurs in the shape of the profile. Such profile shape variations cannot be captured well by the NDACC retrieval, which simply scales a climatological mean profile (WACCM profile), thus leading to a damped seasonality of the time series due to the reduced column sensitivity in the lower troposphere (Fig. 4). Since the seasonal variation is limited to the troposphere, the damping factor depends on the tropopause pressure relative to the site's surface pressure. We assume the damping factor due to the fixed a priori as

$$d_{\text{NDACC}} \sim \frac{1}{d_h}. \quad (5)$$

Here,  $d_h$  is calculated according to Eq. (3).

The central panel of Fig. 7 plots the relative seasonal variation of the NDACC data multiplied by  $d_h$  versus the relative seasonal variations of the TCCON data. We get a good linear correlation ( $R = 0.88$ ) and a linear regression line with



**Figure 10.** Time series of the differences of the yearly means between measurement and respective model. Top: NDACC-model<sub>NDACC</sub>, bottom: TCCON-model.

a slope of 0.43. This indicates that a single damping factor  $d_{\text{NDACC}} = 0.43 \times \frac{1}{d_h}$  is sufficient for describing the damped seasonal variations at all NDACC/FTIR sites. A detailed overview on the damped seasonal cycles for the different sites is given in the Appendix (Fig. A2). Note:  $d_h$  cancels out in Eq. 2 and  $d_{\text{NDACC}} \times d_h$  becomes a constant factor.

## 5 XCO<sub>2</sub> as global proxy for long-term consistency

In this section, we check the NDACC/FTIR time series for long-term consistency by comparing the NDACC XCO<sub>2</sub> measurements with the XCO<sub>2</sub> model calculations.

### 5.1 Comparison of modelled XCO<sub>2</sub> and NDACC XCO<sub>2</sub>

The reduced seasonality due to the fixed a priori used for the NDACC retrievals must be taken into account; i.e. we apply the damping factor  $d_{\text{NDACC}} = 0.43 \cdot \frac{1}{d_h}$  for our model calculations according to Eq. (2) (hereinafter, we call these model calculations with damped seasonality model<sub>NDACC</sub>). Figure 8 shows the same as Fig. 5, but for the correlation between the NDACC XCO<sub>2</sub> and the model<sub>NDACC</sub> data for the time period 1996–2012. NDACC measurements started many years before the TCCON measurements and there are many more data points in Fig. 8 than in Fig. 5. Nevertheless, the correlation to the model is almost as good for NDACC as for TCCON. For monthly mean data (left panel of Fig. 8) we get a correlation coefficient  $R$  of 0.99 and a scatter of 3.5 ‰. For the de-seasonalised yearly mean data (right panel), the scatter is even as small as 2.7 ‰. In agreement with the NDACC vs. TCCON comparison, we observe a systematic positive bias in the NDACC XCO<sub>2</sub> data of about 25 ‰. The damped

seasonality of the NDACC time series is well captured by the model<sub>NDACC</sub> data (central panel of Fig. 8). This documents that the model can also evaluate the NDACC data consistency on intra-annual time scales.

As the subset of NDACC/FTIR sites we are using within this study is representative of nearly all latitudes (Table 4), we conclude that the model<sub>NDACC</sub> is valid for assessing the consistency of all NDACC/FTIR measurements made around the globe.

### 5.2 Network-wide long-term stability

Finally, to document the network-wide long-term stability, we compare annual means, not to be mistaken for the de-seasonalised annual means shown in the comparisons Fig. 5 to Fig. 8. Figure 9 compares the measurements and their respective model with NDACC vs. model<sub>NDACC</sub> on the left and TCCON vs. model on the right. Figure 10 shows the same data, but plotted as a function of time. All these comparisons show that the agreement between measurement and respective model is excellent. There's no drift in the differences, which might be expected when using one fixed a priori over several years. Long-term and station-to-station variabilities of the measurements with respect to the model are within 1 ‰ for both the NDACC and TCCON data sets. The scatter is within 3 ‰ for NDACC-model<sub>NDACC</sub> and 2 ‰ for TCCON-model.

## 6 Conclusions

Mid-infrared high resolution solar absorption spectra have been recorded for many years and at many sites around the globe. Most of these activities are organised within the NDACC and have a high potential for investigating the long-term change of our atmosphere on a global scale. However, such investigations require data that are very consistent throughout many years and between the different sites.

In this work, we present a method that allows an assessment of the consistency of any mid-infrared high-resolution solar absorption measurement (2600–3000 cm<sup>-1</sup> spectral region) made since the late 1950s. The method uses the difference between XCO<sub>2</sub> retrieved from the spectra and as simulated by a model. Both the retrieval and the model are designed in a way that allows their easy adoption to any measurement site.

The XCO<sub>2</sub> retrieval uses a fixed CO<sub>2</sub> a priori profile (obtained from WACCM simulations), which is scaled during the retrieval process. Furthermore, we use NCEP temperature and pressure profiles. This simple scaling retrieval setup using a time-independent a priori allows it to be easily adopted for any site around the globe and for any mid-infrared high-resolution solar absorption measurement if reliable data for the surface pressure and analysis temperature profiles are available. The XCO<sub>2</sub> model is driven by Mauna Loa data (long-term evolution) and CarbonTracker results (latitudinal gradients) and should thus capture well the period covered by the Mauna Loa CO<sub>2</sub> record, which starts in the 1950s.

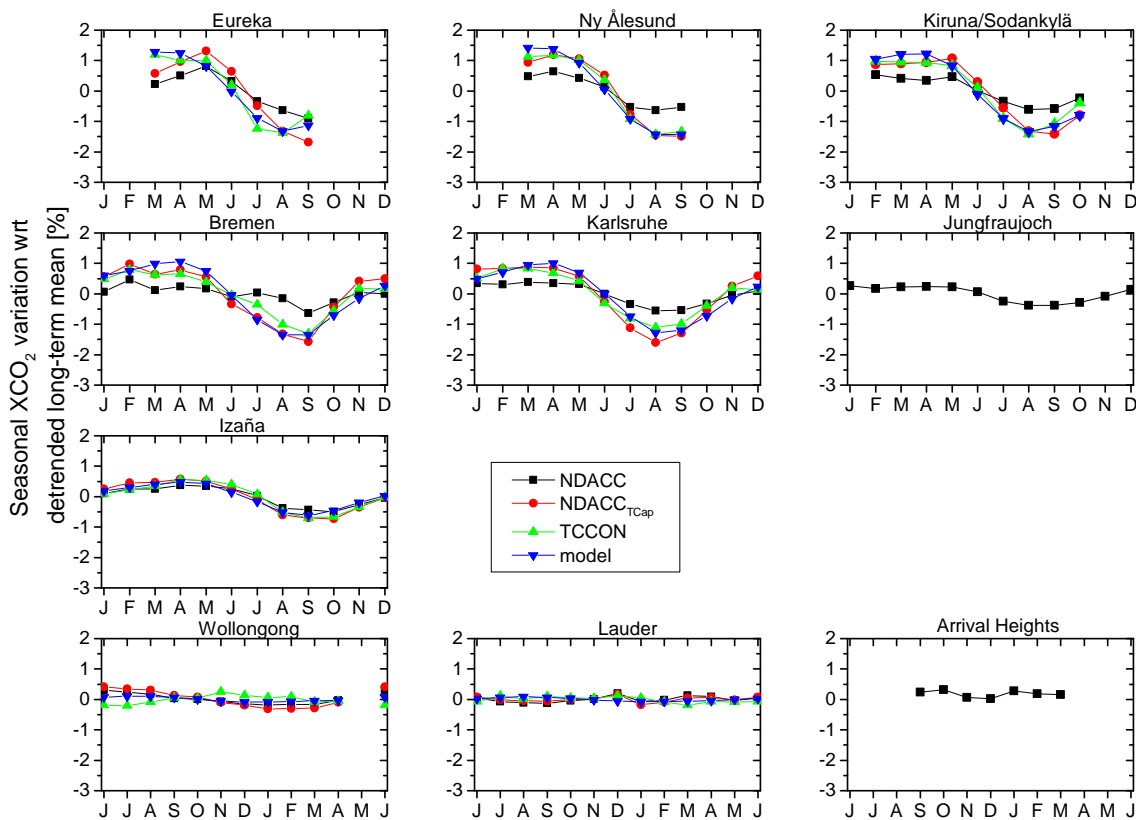
We use the TCCON XCO<sub>2</sub> data to empirically demonstrate the good quality of the NDACC XCO<sub>2</sub> product and of the XCO<sub>2</sub> model simulations. For the period where TCCON data are available, the scatter for monthly mean data between TCCON and model is 2.7 ‰ and between TCCON and NDACC it is 4.1 ‰ (when using the same a priori). We identify a clear systematic difference between the TCCON and NDACC data of 25 ‰ (our NDACC product overestimates the calibrated TCCON values), which is very likely due to an error in the MIR spectroscopic CO<sub>2</sub> parameters. Identifying this clear bias is a side aspect of our study. The important metric we are interested in is the stability of the difference. Furthermore, we demonstrate that using a fixed a priori instead of a daily varying a priori almost exclusively affects the amplitude of the seasonal variations. We show that the respective damping of the seasonal amplitude can be described by a consistent parametrisation for all the different sites and can be very easily considered in the model simulations.

We apply the developed method to the NDACC/FTIR spectra that have so far been contributing to the project MUSICA. These spectra have been measured since 1996 at 10 stations that are distributed around the globe. We found a scatter between the yearly mean NDACC data and the model of about 3 ‰. This provides strong evidence for the very good long-term data consistency between these NDACC/FTIR sites and is a good reliability and consistency test for the long-term trends of tropospheric species measured at these sites.

### Appendix A: Seasonal cycles as determined from the different data sets

All seasonal cycles determined from the different data sets are plotted in Fig. A1. For better comparability, only the subset of measurements is considered, with both, NDACC and TCCON measurements at the same day. Black represents the NDACC values, calculated with fixed a priori information, red are the NDACC values, when the TCCON a priori information is used, green is the seasonal variation of the TCCON time series and blue are the modelled values. It is obvious that the fixed a priori information (black symbols) leads to a reduced sensitivity for the seasonal cycle.

To consider this, the damping factor  $d_{\text{NDACC}}$  is implemented (Sect. 4). The comparison of the detrended seasonal cycle of the whole NDACC data set (black) with the damped model version (red) is shown in Fig. A2. Both are in very good agreement.



**Figure A1.** Seasonal cycles. Black: NDACC with fixed WACCM a priori, red: NDACC with same a priori as TCCON, green: TCCON and blue: model.

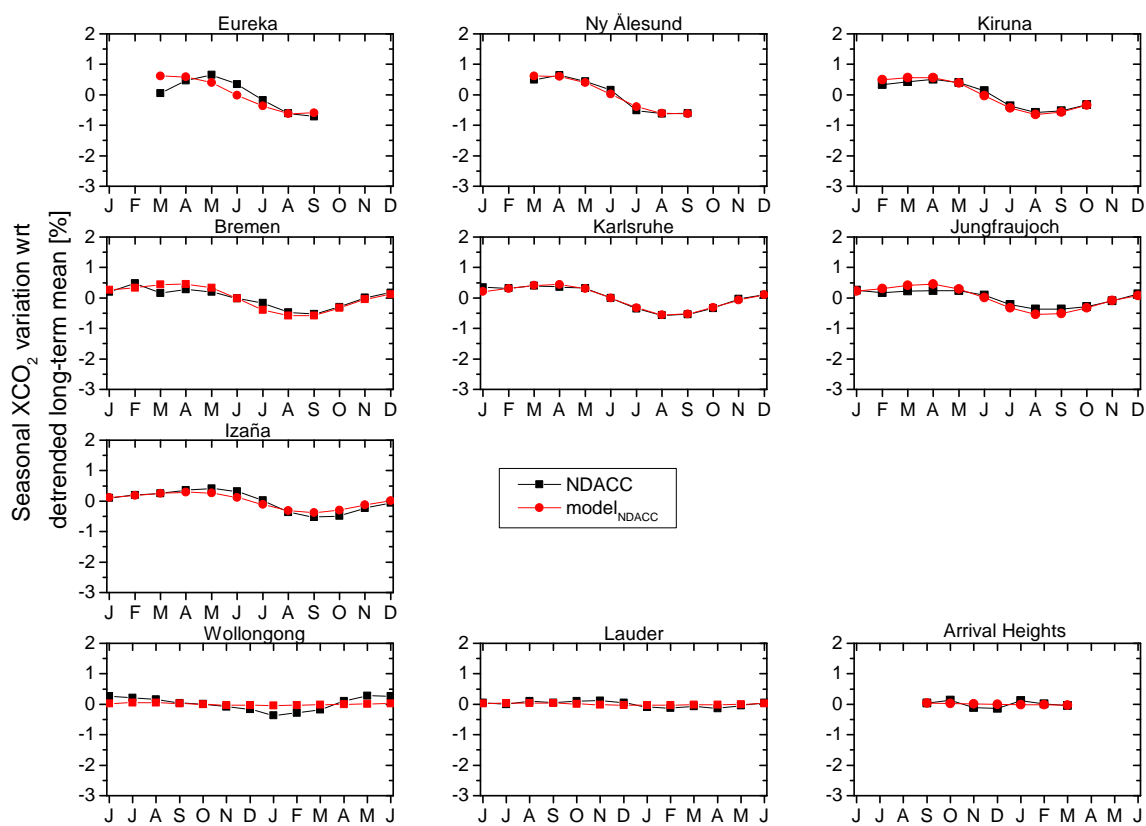


Figure A2. As Fig. A1, but now only NDACC and the damped model (model<sub>NDACC</sub>) are compared.

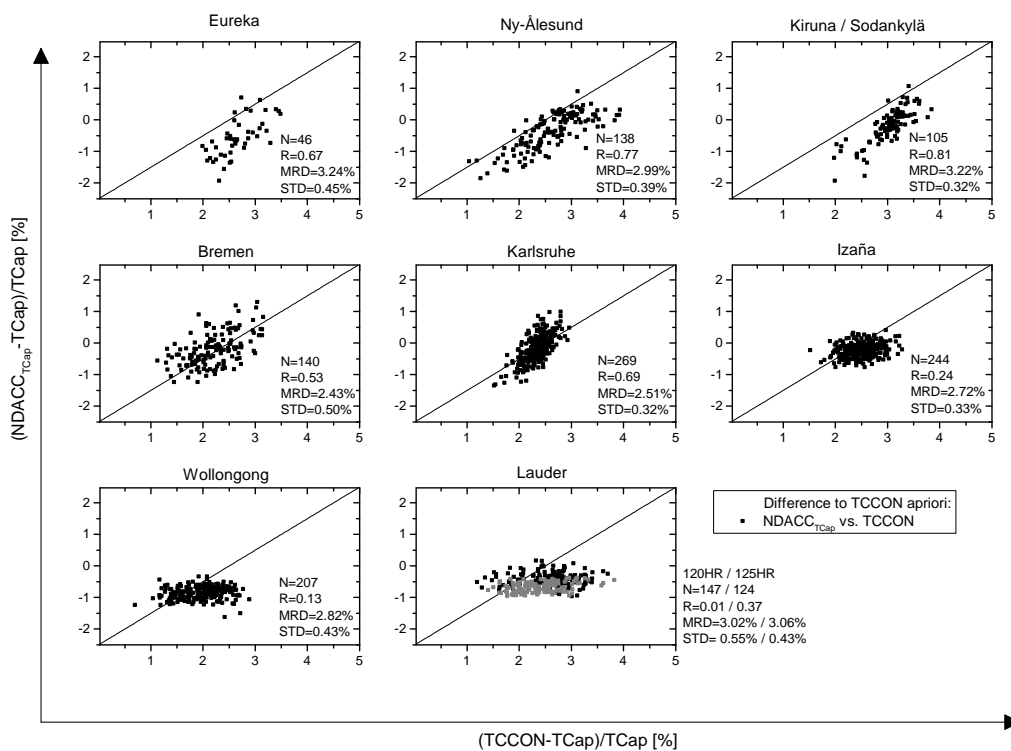


## Appendix B: Comparison of day-to-day XCO<sub>2</sub> variations between TCCON and NDACC

Depicted in Fig. B1 is the correlation of the difference between NDACC (with TCCON a priori, NDACC<sub>Tcap</sub>) and TCCON a priori vs. the difference between TCCON and TCCON a priori. Plotted are the daily means with the number of considered days  $N$ , correlation coefficient  $R$ , mean relative difference (MRD) and standard deviation (SD). The black line is the one-to-one correlation shifted by 2.5 %. By subtracting the a priori, we compare in this plot directly the information that comes from the measurements and compare the capabilities of NDACC and TCCON in improving the XCO<sub>2</sub> estimations as provided by the TCCON a priori model. In the case of Lauder, two TCCON instruments have been used (Bruker 120HR: black points, 125HR: grey points).

There is a higher correlation for northern sites, where there is more variability ( $R$  between 0.5 and 0.7). The scatter values (SD) are between 3 to 5 %. As mentioned before (Sects. 4 and 5), there's a clear systematic difference between the two data. Besides the systematic difference, the agreement of both data sets is good and demonstrates that in the MIR XCO<sub>2</sub> can be obtained at a very good quality (the NDACC measurements improve the empirical TCCON a priori model in a similar way as the TCCON measurements).

We would like to note that the consistency of NDACC and TCCON XCO<sub>2</sub> products might even be further improved by using the same retrieval software and consistent line parameters. The here compared NDACC product works with the TCCON a priori model, but for the retrievals we use the PROFFIT software and the HITRAN spectroscopy (instead of the GGG software with empirical extensions of G. Toon (personal communication, 2014) for the spectroscopy used for the TCCON retrieval).



**Figure B1.** Correlation between  $(\text{NDACC}_{\text{TCap}} - \text{TCap})$  and  $(\text{TCCON} - \text{TCap})$ . The black line represents the one-to-one correlation shifted by 2.5%. The grey squares in the Lauder graph represent the data measured with the 125HR instrument, whereas the black squares are the data measured with the 120HR instrument.

**Appendix C: List of ML<sub>corr</sub> values**

Listed in Table C1 are all yearly correction factors used in our XCO<sub>2</sub> model (Sect. 3). The correction factors ML<sub>corr</sub>(*i*) express the year-to-year variation between the Mauna Loa series compared to a polynomial fit, where *i* is the respective year. For all other years, ML<sub>corr</sub>(*i*) is set to 0. Values within 2 years are calculated by linear interpolation.

**Table C1.** Overview of all yearly correction factors used in our XCO<sub>2</sub> model (Sect. 3).

ML <sub>corr</sub> (1959) = 0.813	ML <sub>corr</sub> (1973) = -0.405	ML <sub>corr</sub> (1987) = 0.946	ML <sub>corr</sub> (2001) = -0.624
ML <sub>corr</sub> (1960) = 0.785	ML <sub>corr</sub> (1974) = -0.509	ML <sub>corr</sub> (1988) = 1.235	ML <sub>corr</sub> (2002) = -0.546
ML <sub>corr</sub> (1961) = 0.677	ML <sub>corr</sub> (1975) = -0.666	ML <sub>corr</sub> (1989) = 1.331	ML <sub>corr</sub> (2003) = -0.303
ML <sub>corr</sub> (1962) = 0.490	ML <sub>corr</sub> (1976) = -0.768	ML <sub>corr</sub> (1990) = 1.141	ML <sub>corr</sub> (2004) = -0.098
ML <sub>corr</sub> (1963) = 0.180	ML <sub>corr</sub> (1977) = -0.633	ML <sub>corr</sub> (1991) = 0.653	ML <sub>corr</sub> (2005) = 0.074
ML <sub>corr</sub> (1964) = -0.119	ML <sub>corr</sub> (1978) = -0.376	ML <sub>corr</sub> (1992) = 0.056	ML <sub>corr</sub> (2006) = 0.134
ML <sub>corr</sub> (1965) = -0.380	ML <sub>corr</sub> (1979) = -0.104	ML <sub>corr</sub> (1993) = -0.442	ML <sub>corr</sub> (2007) = 0.097
ML <sub>corr</sub> (1966) = -0.508	ML <sub>corr</sub> (1980) = 0.138	ML <sub>corr</sub> (1994) = -0.651	ML <sub>corr</sub> (2008) = 0.016
ML <sub>corr</sub> (1967) = -0.566	ML <sub>corr</sub> (1981) = 0.294	ML <sub>corr</sub> (1995) = -0.703	ML <sub>corr</sub> (2009) = -0.061
ML <sub>corr</sub> (1968) = -0.538	ML <sub>corr</sub> (1982) = 0.403	ML <sub>corr</sub> (1996) = -0.619	ML <sub>corr</sub> (2010) = -0.053
ML <sub>corr</sub> (1969) = -0.503	ML <sub>corr</sub> (1983) = 0.481	ML <sub>corr</sub> (1997) = -0.535	ML <sub>corr</sub> (2011) = -0.073
ML <sub>corr</sub> (1970) = -0.537	ML <sub>corr</sub> (1984) = 0.537	ML <sub>corr</sub> (1998) = -0.373	ML <sub>corr</sub> (2012) = -0.064
ML <sub>corr</sub> (1971) = -0.542	ML <sub>corr</sub> (1985) = 0.580	ML <sub>corr</sub> (1999) = -0.386	
ML <sub>corr</sub> (1972) = -0.506	ML <sub>corr</sub> (1986) = 0.702	ML <sub>corr</sub> (2000) = -0.518	

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