

TCCON and NDACC X_{CO} measurements: difference, discussion and application

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Abstract. Column-averaged dry-air mole fraction of CO (X_{CO}) measurements are obtained from two ground-based Fourier transform infrared (FTIR) spectrometers networks: the Total Carbon Column Observing Network (TCCON) and the Network for the Detection of Atmospheric Composition Change (NDACC). In this study, the differences between the TCCON and NDACC X_{CO} measurements are investigated and discussed based on six NDACC/TCCON sites using data over the period 2007-2017. A direct comparison shows that the NDACC X_{CO} measurements are about 5.5% larger than the TCCON data at Ny-Ålesund, Bremen, and Izaña (Northern Hemisphere), and the absolute bias between the NDACC and TCCON data are within 2% at St Denis, Wollongong and Lauder (Southern Hemisphere). The hemispheric dependence of the bias is mainly attributed to their smoothing errors. The systematic smoothing error of the TCCON X_{CO} data varies in the range between 0.2% (Bremen) and 7.9% (Lauder), and the random smoothing error in the range between 2.0% and 3.6%. The systematic smoothing error of NDACC data is between 0.1% and 0.8%, and the random smoothing error of NDACC data is about 0.3%. For TCCON data, the smoothing error is significant because it is higher than the reported uncertainty, particularly at Southern Hemisphere sites. To reduce the influence from the a priori profiles and different vertical sensitivities, the scaled NDACC a priori profiles are used as the common a priori profiles for comparing TCCON and NDACC retrievals. As a result, the biases between TCCON and NDACC X_{CO} measurements become more consistent (5.6-8.5%) with a mean value of 6.8% at these sites. To determine the sources of the remaining bias, regular AirCore measurements at Orleans and Sodankylä are compared to co-located TCCON measurements. It is found that TCCON X_{CO} measurements are $6.1 \pm 1.6\%$ and $8.0 \pm 3.2\%$ smaller

than the AirCore measurements at Orleans and Sodankylä respectively, indicating that the scaling factor of TCCON X_{CO} data should be around 1.0000 instead of 1.0672. Further investigations should be carried out in the TCCON community to determine the correct scaling factor to be applied to the TCCON X_{CO} data. This paper also demonstrates that the smoothing error must be taken into account when comparing FTIR X_{CO} data, and especially TCCON X_{CO} data, with model or satellite data.

5 1 Introduction

Carbon monoxide (CO) is a trace gas in the Earth's atmosphere, with a typical mole fraction of 50 - 80 ppb (parts per billion) at clean-air sites. Atmospheric CO is released by incomplete combustion, mainly coming from anthropogenic emissions (Granier et al., 2011), and biomass burning (van der Werf et al., 2010). There are also small quantities of CO in the mesosphere generated by the photolysis of carbon dioxide (Garcia et al., 2014). The lifetime of CO is about two months in the troposphere (Pfister et al., 2004), and on the order of several months in the stratosphere (Hoor et al., 2004). CO is often used as a tracer to study the long-distance transport of biomass burning (Dufлот et al., 2010), wildfires (Turquety et al., 2009) and anthropogenic emissions (Ojha et al., 2016). The major sink of CO in the atmosphere is the reaction with hydroxyl radicals (OH) (Spivakovsky et al., 2000). Therefore, CO plays an important role in atmospheric chemistry and thus affects the atmospheric oxidizing capacity. CO concentration is associated with many tropospheric polluting gases, e.g., tropospheric ozone and urban smog (Aschi and Largo, 2003), and it also has a strong impact on the carbon and methane cycles (Rasmussen and Khalil, 1981).

Global CO total columns are measured by space-based satellite instruments, e.g. the measurement of pollution in the troposphere (MOPITT), the scanning imaging absorption spectrometer for atmospheric cartography (SCIAMACHY), the infrared atmospheric sounding interferometer (IASI) and the more recently tropospheric monitoring instrument (TROPOMI) (Deeter et al., 2017; Borsdorff et al., 2016; George et al., 2009; Borsdorff et al., 2018). Satellite measurements are applied to study the long-term trend of CO (Worden et al., 2013), to understand the regional pollution (Dekker et al., 2019) and are assimilated into the atmospheric chemistry model to improve air quality forecasts (Klonecki et al., 2012; Mizzi et al., 2016). To better understand the uncertainties of the satellite CO observations and the model simulations, they need to be validated by other measurements. Ground-based Fourier transform infrared (FTIR) spectrometers record the direct solar radiation and observe the total column of CO with high accuracy and precision. In addition, the ground-based FTIR CO measurements are stable over a long-time period, so that they can be used to validate the satellite CO observations (Dils et al., 2006; Borsdorff et al., 2016, 2018) and model simulations (Eskes et al., 2015). Nowadays, there are two well-known global ground-based FTIR networks providing total column-averaged dry air mole fraction of CO (X_{CO}) measurements: the Total Carbon Column Observing Network (TCCON) (Wunch et al., 2011) and the Network for the Detection of Atmospheric Composition Change (NDACC) (De Mazière et al., 2018).

TCCON and NDACC X_{CO} measurements are sometimes combined together to validate satellite observations or model simulations, and it is noticed that the smoothing error of TCCON and NDACC X_{CO} measurements are not always taken into account when comparing with satellite observations, e.g. SCIAMACHY (Borsdorff et al., 2016; Hochstaffl et al., 2018) and TROPOMI (Borsdorff et al., 2018) because it is considered to have a negligible impact. By using both TCCON and

NDACC X_{CO} data to validate the SCIAMACHY observations, Borsdorff et al. (2016) found that NDACC X_{CO} data is 3.8 ppb larger than TCCON measurements. Despite of the similar measurement technique, there are differences between TCCON and NDACC X_{CO} products because the observed spectra, retrieval algorithms and data corrections are different. To understand why there is a systematic bias between the TCCON and NDACC X_{CO} measurements, a case study was carried out by Kiel et al. (2016) using TCCON and NDACC measurements at Karlsruhe during 2010-2014. They found that NDACC X_{CO} is 4.47 ± 0.17 (1σ) ppb larger than the TCCON data, and the difference between the TCCON and NDACC X_{CO} measurements mainly comes from the air mass independent (scaling) correction of the TCCON data, and partly from the air mass dependent correction, the spectroscopic parameters and a priori profiles.

In this study, the comparison between the TCCON and NDACC X_{CO} measurements is extended to six sites (Ny-Ålesund, Bremen, Izaña, St Denis, Wollongong and Lauder) during the time period of 2007-2017. This work aims at understanding (1) whether the bias between TCCON and NDACC X_{CO} measurements is consistent at these sites, (2) whether the smoothing uncertainties of TCCON and NDACC X_{CO} measurements can be ignored when comparing against each other or other datasets, and (3) whether the scaling factor of TCCON X_{CO} data is correct. This paper is organized as follows. Section 2 lists the FTIR sites used in this study and describes the main characteristics of the TCCON and NDACC X_{CO} measurements. Direct comparisons between TCCON and NDACC X_{CO} measurements are carried out in Section 3. In Section 4, the differences between TCCON and NDACC X_{CO} measurements are investigated in relation to their a priori profiles and averaging kernels. The smoothing errors of TCCON and NDACC X_{CO} measurements are estimated. The TCCON X_{CO} measurements are compared with AirCore measurements at Sodankylä and Orleans. Section 5 shows an example of using TCCON and NDACC X_{CO} measurements together in a comparison with a model simulation. Conclusions are drawn in Section 6.

2 FTIR measurements

The ground-based FTIR measurement system is composed of an automatic weather station, a sun tracker and a FTIR instrument. The locations of the FTIR sites used in this study and time coverages of the TCCON and NDACC X_{CO} measurements are listed in Table 1. All these sites use a Bruker IFS 120/125HR instrument to record near infrared (NIR) spectra for TCCON measurements and mid infrared (MIR) spectra for NDACC measurements. The main characteristics of TCCON and NDACC X_{CO} measurements are described below.

2.1 TCCON

TCCON uses the GGG2014 code that applies a profile scaling to retrieve CO and O₂ total columns simultaneously (Wunch et al., 2015). The spectral resolution of the NIR spectrum is 0.02 cm^{-1} . The retrieval windows of CO are $4208.7\text{-}4257.3 \text{ cm}^{-1}$ and $4262.0\text{-}4318.8 \text{ cm}^{-1}$. The interfering species are CH₄, H₂O and HDO. The retrieval window of O₂ is $7765.0\text{-}8005.0 \text{ cm}^{-1}$, with interfering absorptions from H₂O, HF, CO₂ and solar lines. The spectroscopy is the atmospheric line list (ATM) linelist maintained at Jet Propulsion Laboratory, NASA (Toon, 2014). Since the O₂ volume mixing ratio (VMR) of 0.2095 is constant in the atmosphere, TCCON uses the O₂ total column (TC_{O_2}) to calculate the total column of the dry air

Table 1. The coordinates, responsible institute and time coverage of measurements at six sites used in this study.

Site	Latitude	Longitude	Altitude (km a.s.l)	Research group	Time coverage (TCCON/NDACC)	Instrument
Ny-Ålesund	78.9°N	11.9°E	0.02	U. of Bremen	2007-2017/2007-2017	Bruker 120HR
Bremen	53.1°N	8.8°E	0.03	U. of Bremen	2009-2017/2007-2016	Bruker 125HR
Izaña	28.3°N	16.5°W	2.37	AEMET & KIT	2007-2017/2007-2017	Bruker 125HR
St Denis (Reunion Island)	21.0°S	55.4°E	0.08	BIRA-IASB	2011-2017/2011-2015	Bruker 125HR
Wollongong	34.4°S	150.9°E	0.03	U. of Wollongong	2008-2017/2008-2017	Bruker 125HR
Lauder	45.0°S	169.7°E	0.37	NIWA	2010-2017/2007-2017	Bruker 120/5HR

($TC_{dry,air} = TC_{O_2}/0.2095$), and then to calculate the X_{CO} as the ratio between the retrieved CO total column (TC_{CO}) and the total column of the dry air ($X_{CO} = 0.2095 \times \frac{TC_{CO,r}}{TC_{O_2,r}}$). Furthermore, TCCON X_{CO} data have been indirectly validated by several aircraft and AirCore measurements, and the publicly available TCCON X_{CO} data have been corrected with a scaling factor (α) and an airmass dependent factor (β) (Wunch et al., 2015)

$$5 \quad X_{CO} = 0.2095 \times \frac{TC_{CO,r}}{TC_{O_2,r}} \times \frac{1}{\alpha \cdot [1 + \beta \times SBF(\theta)]}, \quad (1)$$

where $\alpha = 1.0672$ and $\beta = -0.0483$, θ is the solar zenith angle (SZA) and the $SBF(\theta)$ depends on the probed airmass through the SZA ($SBF(\theta) = [(\theta + 13)/(90 + 13)]^3 - [(45 + 13)/(90 + 13)]^3$).

According to Figure 10 in Wunch et al. (2015), the random uncertainty of TCCON X_{CO} data is below 3.5% and decreases with increasing SZA. The largest source is the uncertainty of the observer-sun Doppler stretch (osds) due to a solar tracker pointing uncertainty. The shear misalignment, continuum curvature and a priori profile shape are the other leading sources of uncertainty, and they are all about 1.0%. In this study, it is assumed that the mean random uncertainty of TCCON X_{CO} measurement is 3.5% as a upper limitation. Since TCCON data have been scaled to the WMO standard, the systematic uncertainty of TCCON X_{CO} data is eliminated and it is assumed to be zero. Note that the systematic smoothing error has not been removed in public TCCON data, because the Aircraft or AirCore profiles which are used to calibrate the TCCON X_{CO} measurements have been first smoothed with TCCON data (Wunch et al., 2010).

2.2 NDACC

NDACC uses either the SFIT4 (Pougatchev et al., 1995) or the PROFFIT9 code (Hase et al., 2004) to retrieve CO vertical profiles. The retrieval windows for CO are 2057.70-2058.00 cm^{-1} , 2069.56-2069.76 cm^{-1} and 2157.50-2159.15 cm^{-1} . The spectral resolution of the MIR spectrum is about 0.0035 - 0.0070 cm^{-1} . The interfering species are O₃, CO₂, OCS, N₂O and H₂O. The reference spectroscopy database is HITRAN2008 (Rothman et al., 2009). Since the O₂ total column is not available from the NDACC spectrum and the weak N₂ (a potential alternative) signal in the NDACC region leads to a large scatter, the total column of the dry air is computed from the surface pressure (P_s) recorded at a local automatic weather station and the

National Centers for Environmental Prediction (NCEP) reanalysis H₂O total column (TC_{H_2O})

$$X_{CO} = \frac{TC_{CO,r}}{TC_{dry,air}} = \frac{TC_{CO,r}}{P_s / (gm_{air}^{dry}) - TC_{H_2O} (m_{H_2O} / m_{air}^{dry})}, \quad (2)$$

where g is the column-averaged gravity acceleration, m_{H_2O} and m_{air}^{dry} are the molecular masses of H₂O and dry air respectively. Unlike TCCON X_{CO} data, there are no scaling nor airmass dependent corrections for NDACC data.

- 5 The NDACC X_{CO} data is calculated by the ratio between the total column of CO and the total column of the dry air. Zhou et al. (2018) pointed out that the uncertainty of the total column of the dry air is within 0.1% by using the surface pressure and NCEP water vapor. Therefore, the uncertainty of the NDACC X_{CO} data is dominated by the uncertainty of the retrieved total column of CO. To understand the error budget for NDACC CO data, the different contributions to the total uncertainty budget at St Denis are listed in Table 2. The systematic uncertainty is mainly coming from the spectroscopic parameters and
- 10 temperature profile, while the random uncertainty is mainly coming from the SZA and temperature. Note that the systematic and random smoothing errors are not included in the reported NDACC data. The uncertainty of NDACC CO total column data can be variable, depending on site-specific conditions, e.g. humidity, instrument, location and retrieval software (see Table 3).

Table 2. The systematic and random uncertainties for NDACC retrieved X_{CO} at St Denis. '-' means that the uncertainty is less than 0.1 and then can be ignored. The "Total" uncertainties are calculated by adding the sub-types in quadrature.

	Systematic [%]	Random [%]
Measurement	-	0.1
Spectroscopy	2.0	-
SZA	0.1	0.7
Temperature	1.5	0.7
Dry air column	0.1	0.1
Total	2.5	1.0

Table 3. The systematic and random uncertainties of NDACC retrieved CO total column.

Site	Ny-Ålesund	Bremen	Izaña	St Denis	Wollongong	Lauder
sys/ran [%]	4.0/5.0	3.4/4.0	2.1/0.5	2.5/1.0	2.1/2.2	2.1/1.8

3 TCCON and NDACC direct comparisons

- Figure 1 shows the direct comparisons between TCCON and NDACC X_{CO} co-located hourly means at the six sites. The
- 15 TCCON and NDACC measurements observe the same seasonal cycles of X_{CO} . At Northern Hemisphere stations (Ny-Ålesund, Bremen and Izaña), the seasonal variation of X_{CO} is dominated by the OH variation (Té et al., 2016), with a low value of

X_{CO} in the summer (June-August) and a high value in the winter (December-February). At Southern Hemisphere stations (St Denis, Wollongong and Lauder), the seasonal variation of X_{CO} is dominated by biomass burning, with a peak in September-November (Duflot et al., 2010). The correlation coefficients (R) at the six sites are between 0.96 and 0.99, indicating good agreement between TCCON and NDACC X_{CO} measurements.

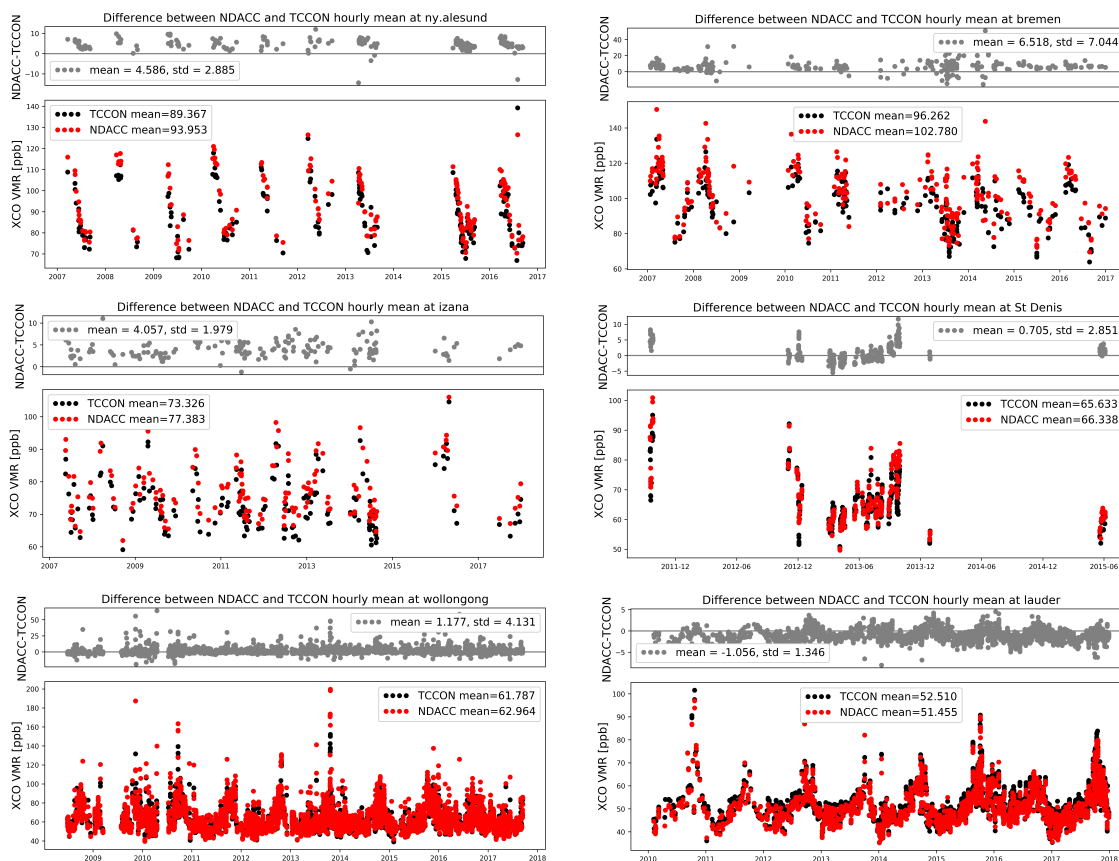


Figure 1. The time series of the TCCON and NDACC X_{CO} measurements, together with their differences in unit of ppb. Note that the range of the y axes is different at each site due to a large variation of CO in the atmosphere.

5 Table 4 shows the relative mean and standard deviation (SD) between the TCCON and NDACC X_{CO} measurements at these sites. The mean relative biases are about 5.5% at Ny-Ålesund, Bremen and Izaña (Northern Hemisphere), and the absolute bias between the NDACC and TCCON data are within 2% at St Denis, Wollongong and Lauder (Southern Hemisphere). The difference in the mean bias between the two hemispheres is up to 5.2%. Apart from the large SDs of 6.9% and 6.6% at Bremen and Wollongong, respectively, the SDs are quite similar among other sites with a range from 2.6% to 4.3%. According to

10 Rodgers (2003), if we ignore the smoothing error of two datasets, the systematic and random uncertainties of the differences

between standard TCCON and NDACC measurements are calculated as

$$\varepsilon_{sys} = \varepsilon_{sys,N}, \quad (3)$$

$$\varepsilon_{ran} = \sqrt{\varepsilon_{ran,T}^2 + \varepsilon_{ran,N}^2}, \quad (4)$$

where $\varepsilon_{sys,N}$ is the systematic uncertainty of NDACC X_{CO} measurements, $\varepsilon_{ran,T}$, $\varepsilon_{ran,N}$ are the random uncertainties of TCCON and NDACC X_{CO} measurements, respectively. Table 4 shows that the mean bias is higher than the systematic uncertainty at Ny-Ålesund, Bremen and Izaña, while the SD is higher than the random uncertainty at St Denis and Wollongong.

Table 4. The relative mean and SD between the TCCON and NDACC X_{CO} measurements ((NDACC-TCCON)/NDACC \times 100%) at six sites, together with the systematic and random uncertainties of the differences between public TCCON and NDACC measurements. The relative mean and SD between the TCCON and NDACC (with and without correction) X_{CO} measurements using the common optimal a priori profile.

		Ny-Ålesund	Bremen	Izaña	St Denis	Wollongong	Lauder
Direct comparison	mean \pm SD [%]	4.9 \pm 3.1	6.4 \pm 6.9	5.2 \pm 2.6	1.1 \pm 4.3	1.9 \pm 6.6	-2.0 \pm 2.6
	sys/ran [%]	4.0/6.1	3.4/5.3	2.1/3.5	2.5/3.6	2.1/4.1	2.1/3.9
Common a priori profile	mean \pm SD [%]	8.5 \pm 4.2	6.2 \pm 6.8	7.7 \pm 3.2	6.3 \pm 5.1	6.2 \pm 7.6	5.6 \pm 3.5
Common a priori profile but uncorrected TCCON	mean \pm SD [%]	1.5 \pm 4.2	-0.8 \pm 6.8	0.7 \pm 3.2	-0.7 \pm 5.1	-0.8 \pm 7.6	-1.4 \pm 3.5

The ground-based FTIR records the direct solar radiation, and the light path is related to the SZA. Because of the uncertainty from the spectroscopy, the TCCON X_{CO} data have been corrected with an airmass dependent factor (see Eq. 1). No correction is applied to the NDACC data. To check if there is a SZA dependent in the difference between TCCON and NDACC X_{CO} measurements, the differences varying with SZA are shown in Figure 2. Because of the different mean biases, the data are plotted separately in the Northern Hemisphere and in the Southern Hemisphere. In summary, the differences resulting from SZA are very small in both hemispheres, compared to the large scatter.

4 Discussions

In this section, we investigate the causes of the difference between the TCCON and NDACC X_{CO} data. Based on the optimal estimation method (Rodgers, 2000), the TCCON and NDACC retrieved X_{CO} can be written as

$$X_{r,T} = \frac{TC_{r,T}}{\alpha' TC_{air}^{dry}} = \frac{1}{TC_{air}^{dry}} [TC_{a,T} + \mathbf{A}_T(\mathbf{PC}_t - \mathbf{PC}_{a,T})] + \left[\varepsilon_{sys,T} - (1 - 1/\alpha') \frac{TC_{r,T}}{TC_{air}^{dry}} \right] \pm \varepsilon_{ran,T}, \quad (5)$$

$$X_{r,N} = \frac{TC_{r,N}}{TC_{air}^{dry}} = \frac{1}{TC_{air}^{dry}} [TC_{a,N} + \mathbf{A}_N(\mathbf{PC}_t - \mathbf{PC}_{a,N})] + \varepsilon_{sys,N} \pm \varepsilon_{ran,N}, \quad (6)$$

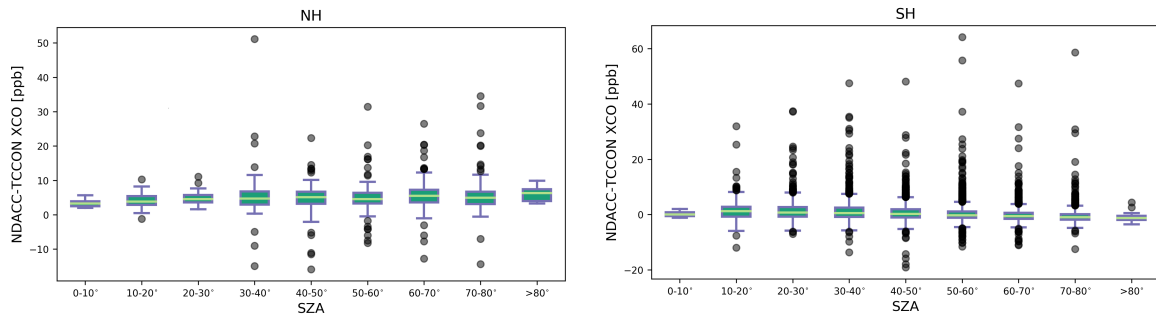


Figure 2. The box plot of the differences between the TCCON and NDACC X_{CO} measurements as a function of SZA for Northern Hemisphere (left) and Southern Hemisphere sites (right). The bottom and upper boundaries of the box represent the 25% and 75% percentile of the data points around their a median value (green line) and the errorbars indicate the 5% and 95% percentiles of the data points.

where, the subscript T and N point to TCCON and NDACC respectively, X_r is the retrieved X_{CO} , TC_a is the a priori total column of CO, A is the column average kernel, PC_t and PC_a are the true and the a priori partial column profiles respectively and ε is the uncertainty. Note that, the $\varepsilon_{sys,T}$ and $\varepsilon_{ran,T}$ are the systematic and random uncertainties of the uncorrected TCCON data (without scaling correction, air mass dependent correction and using surface pressure to calculate the dry air column). α' represents the calculation of the dry air column and air mass independent and air mass dependent corrections in the TCCON procedure. The systematic uncertainty of the corrected TCCON data (standard product) is eliminated by its processing ($[\varepsilon_{sys,T} - (1 - 1/\alpha') \frac{TC_{r,T}}{TC_{air}^{dry}}] = 0$). It is assumed that the random uncertainty is not affected by the α' , as α' is close to 1.0 and the first order of the random uncertainty is unchanged. α' is calculated as

$$\alpha' = \alpha \cdot \overline{TC_{O_2} / (0.2095 TC_{dry,air})} \cdot [1 + \overline{\beta \times SPF(\theta)}] = 1.076, \quad (7)$$

where $\alpha = 1.0672$ ($1\sigma : 0.0200$) is the scaling factor in the GGG2014 code, $\overline{TC_{O_2} / (0.2095 TC_{dry,air})} = 1.016$ ($1\sigma : 0.002$) is the difference in the dry air total column between the O_2 column and surface pressure, $[1 + \overline{\beta \times SPF(\theta)}] = 0.992$ ($1\sigma : 0.003$) is the air mass dependent correction. We calculate $\overline{TC_{O_2} / (0.2095 TC_{dry,air})}$ and $[1 + \overline{\beta \times SPF(\theta)}]$ based on the TCCON measurements at these six sites.

The difference between the standard TCCON and NDACC X_{CO} measurements can then be written as

$$X_{r,N} - X_{r,T} = \frac{1}{TC_{air}^{dry}} ([TC_{a,N} + A_N(PC_t - PC_{a,N})] - [TC_{a,T} + A_T(PC_t - PC_{a,T})]) + \varepsilon_{sys,N} \pm \sqrt{\varepsilon_{ran,N}^2 + \varepsilon_{ran,T}^2}. \quad (8)$$

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Apart from the retrieval uncertainties, the difference between the TCCON and NDACC X_{CO} data also includes the impact from the different a priori profiles and averaging kernels of TCCON and NDACC measurements. The a priori profile of TCCON is generated on a daily basis by the GGG2014 code (Toon and Wunch, 2014), based on Mark IV balloon interferometer (MkIV) and Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS) profiles measured in the 30-40°N latitude range from 2003 to 2007 and taking into account the tropopause height variation and the secular trend. The mean of

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the monthly means during 1980-2020 from the Whole Atmosphere Community Climate Model (WACCM) version 6 is used as the a priori profile for the NDACC retrievals (constant in time) at Ny-Ålesund, Bremen, Izaña, St Denis and Wollongong. The a priori profile for NDACC retrievals at Lauder is constructed from several Atmospheric Trace Molecule Spectroscopy (ATMOS) and aircraft observations. The CO a priori profiles of TCCON and NDACC measurements at these six sites are shown in Figure 3. The TCCON and NDACC a priori profiles are very different. The TCCON a priori profiles at the six sites are close to each other in the stratosphere, which is due to the fact that the stratospheric part of TCCON a priori profile is mainly generated based on the MkIV and ACE-FTS profiles measured in the 30-40°N latitude range. The TCCON a priori profiles in the troposphere at Ny-Ålesund, Bremen and Izaña are close to each other, and are very different with those at St Denis, Wollongong and Lauder. The NDACC CO a priori profiles are much more variable than TCCON a priori profiles both in the troposphere and in the stratosphere. Based on previous studies and emission inventories, the a priori profile shapes from NDACC seem to be more realistic. For example, at St Denis, the CO VMR in the middle and upper troposphere is much larger than that in the lower troposphere, because the air in the lower altitude is relatively clean, coming mainly from the Indian Ocean, while the air mass in the middle and upper troposphere is more polluted coming mainly from Africa and South America (Dufлот et al., 2010; Zhou et al., 2018). At Bremen, the CO VMR in the boundary layer is much larger than the CO VMR in the free troposphere, because there are strong local anthropogenic emissions (European Commission, 2013).

The column averaging kernels (AVKs) of TCCON and NDACC retrievals are different due to their different retrieval windows, spectral resolution and retrieval settings. The AVKs of TCCON and NDACC retrievals at St Denis are shown in Figure 4. In general, the TCCON column AVK increases with altitude which implies that TCCON retrieved CO total column tends to underestimate a deviation from the a priori profile in the troposphere and to overestimate a deviation from the a priori profile in the stratosphere. NDACC exhibits uniform sensitivity in the troposphere and varies in the stratosphere with SZA. As a result, NDACC retrieved CO total columns correctly capture a deviation from the a priori partial column in the troposphere and generally underestimate a deviation from the a priori partial column in the stratosphere.

4.1 Using common a priori profile

To better compare the TCCON and NDACC retrievals, a common optimal a priori profile (subscript *op*) is applied to both TCCON and NDACC retrievals (Rodgers, 2003). The TCCON and NDACC retrieved X_{CO} are

$$X'_{r,T} = \frac{1}{TC_{air}^{dry}} [TC_{op} + \mathbf{A}_T(\mathbf{PC}_t - \mathbf{PC}_{op})] + \left[\varepsilon_{sys,T} - (1 - 1/\alpha') \frac{TC'_{r,T}}{TC_{air}^{dry}} \right] \pm \varepsilon_{ran,T}, \quad (9)$$

$$X'_{r,N} = \frac{1}{TC_{air}^{dry}} [TC_{op} + \mathbf{A}_N(\mathbf{PC}_t - \mathbf{PC}_{op})] + \varepsilon_{sys,N} \pm \varepsilon_{ran,N}, \quad (10)$$

where \mathbf{PC}_{op} is the common a priori partial column profile, TC_{op} is the a priori total column, and $TC'_{r,T}$ is the uncorrected retrieved TCCON CO total column with the optimal a priori profile. The difference between the TCCON and NDACC X_{CO} becomes

$$X'_{r,N} - X'_{r,T} = \frac{(\mathbf{A}_N - \mathbf{A}_T) \cdot (\mathbf{PC}_t - \mathbf{PC}_{op})}{TC_{air,dry}} + \left[\left((1 - 1/\alpha') \frac{TC'_{r,T}}{TC_{air}^{dry}} - \varepsilon_{sys,T} \right) \pm \varepsilon_{sys,N} \right] \pm \sqrt{\varepsilon_{ran,N}^2 + \varepsilon_{ran,T}^2}. \quad (11)$$

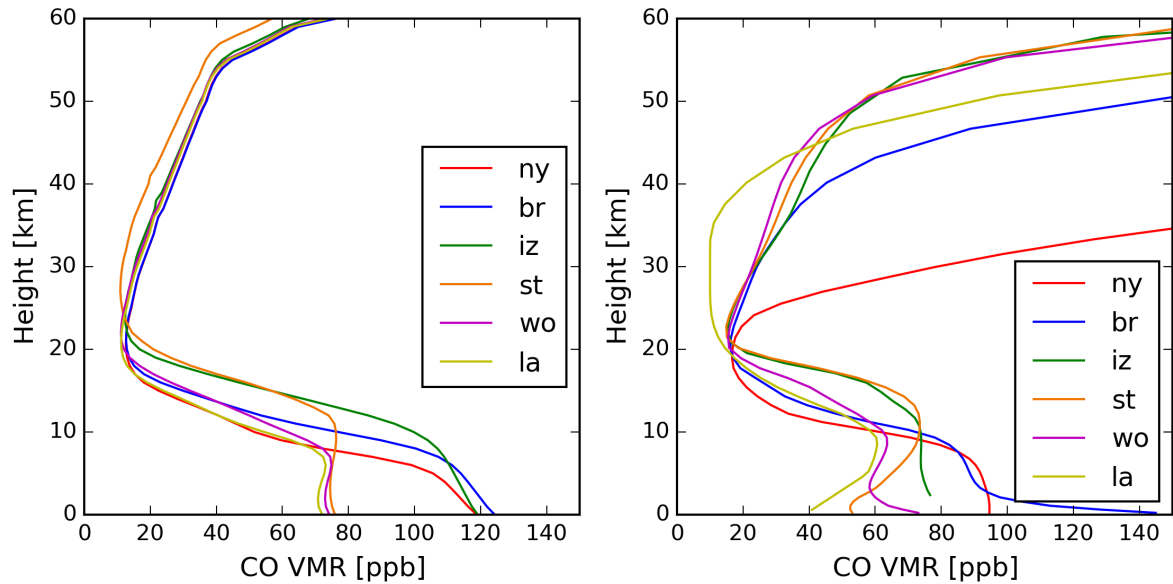


Figure 3. The CO a priori VMR profiles for TCCON (left) and NDACC (right) at six sites (ny: Ny-Ålesund; br: Bremen; iz: Izaña; st: St Denis; wo: Wollongong; la: Lauder). As TCCON a priori profiles change every day, the mean profiles in 2013 are shown here.

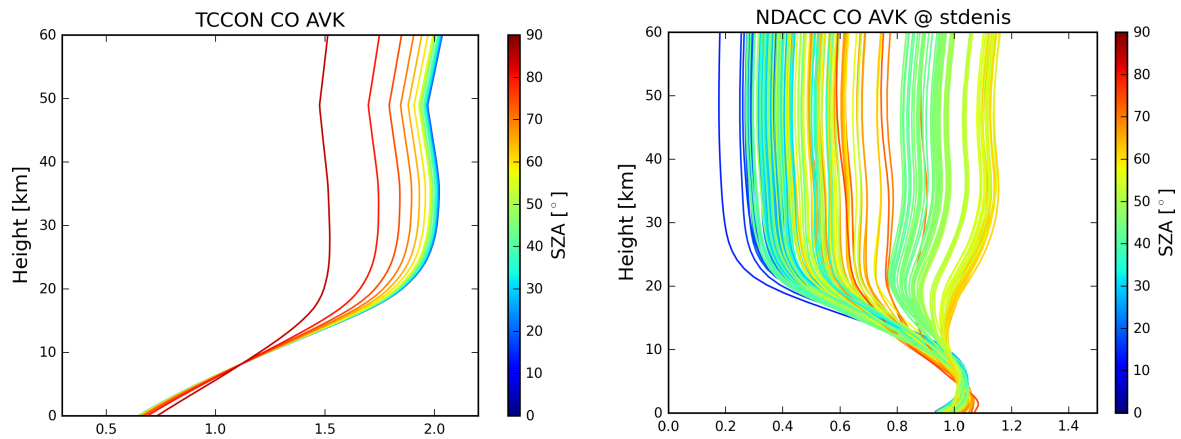


Figure 4. The column averaging kernels of TCCON (left) and NDACC (right) CO retrievals at St Denis.

We keep the systematic uncertainty here, in case the correction of the TCCON data does not get rid of the systematic uncertainty completely. If the optimal common a priori profile is close to the true status, then the first item in the right-band side of the Eq.

11 can be neglected and the difference between the TCCON and NDACC X_{CO} data becomes

$$X'_{r,N} - X'_{r,T} \approx [((1 - 1/\alpha')X_{op} - \varepsilon_{sys,T}) \pm \varepsilon_{sys,N}] \pm \sqrt{\varepsilon_{ran,N}^2 + \varepsilon_{ran,T}^2}, \quad (12)$$

where $(1 - 1/\alpha') = 0.070$ and $X_{op} = TC_{op}/TC_{air}^{dry}$. There is a systematic (constant) difference between the TCCON and NDACC X_{CO} products of about 7.0%, because of the airmass correction, airmass independent correction and the method of calculating dry air column of TCCON data.

Figure 5 shows the TCCON a priori and retrieved TCCON profiles, together with NDACC a priori and scaled NDACC a priori profiles along with HIPPO CO measurements at Wollongong and Lauder. For the scaled NDACC a priori profile, the scaling factor is calculated as the ratio between each retrieved NDACC CO total column and a priori CO total column ($x_{N,scaled} = x_{N,ap} \times TC_{N,r}/TC_{N,ap}$). By comparing against HIPPO measurements, it is found that the vertical variability in TCCON a priori profile is too small and both the TCCON and NDACC a priori profiles have systematic biases. In summary, the scaled NDACC a priori profile is the most reasonable a priori profile among them. Instead of using another model profile, which is not always available to the TCCON and NDACC data users, we chose scaled NDACC a priori profiles as the common a priori profiles for TCCON and NDACC measurements.

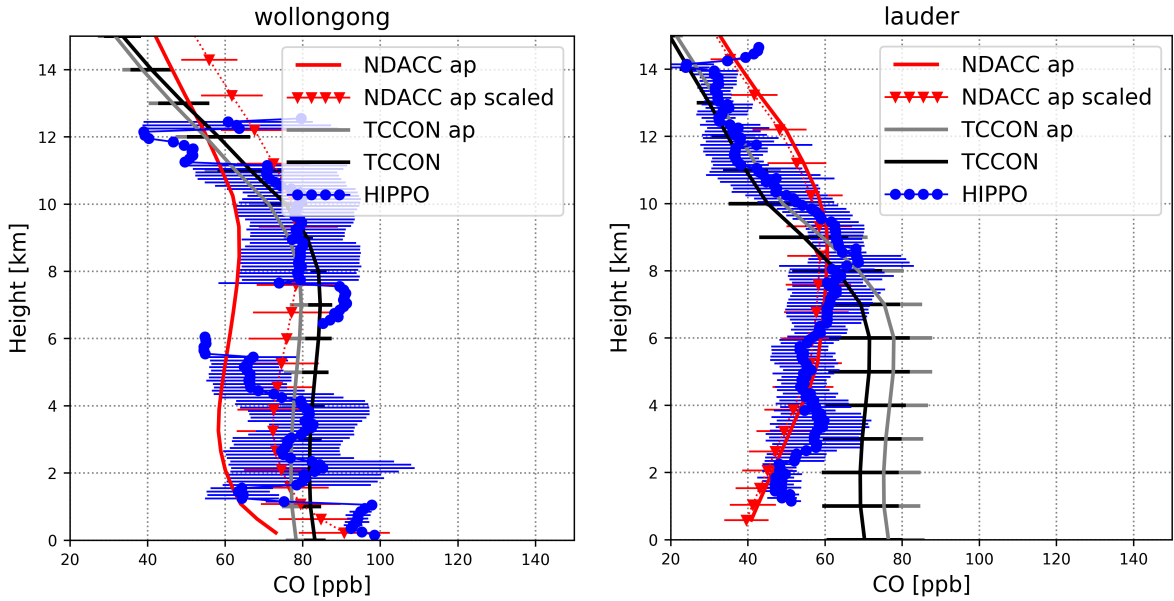


Figure 5. The vertical distribution of NDACC a priori profile (NDACC ap), scaled NDACC a priori profiles (NDACC ap scaled), TCCON a priori profiles (TCCON ap), TCCON retrieved profiles (TCCON) and HIPPO aircraft measurements (HIPPO) in the range from surface to 15 km at Wollongong (left) and Lauder (right). The errorbar is the SD for each dataset.

The systematic smoothing error is reduced by using the updated a priori profile. The differences between the TCCON and NDACC X_{CO} measurements by using the scaled NDACC a priori profile as the common a priori profile are also listed in the

Table 4. The biases become 5.6% to 8.5% with a mean value of 6.8%, and there is almost no inter-hemispheric dependence. However, the bias is beyond the systematic uncertainty at all sites. If we use the uncorrected TCCON data (scaling TCCON data by +7% according to Eq. 12; see Table 4), then the differences between the TCCON and NDACC X_{CO} measurements at these sites become -1.4 - 1.5%. It seems that the processing and correction of the TCCON data, especially the scaling factor, leads into the bias, which is consistent with the result of Kiel et al. (2016).

4.2 Smoothing error estimation

Although the scaled NDACC a priori profile seems to be a good candidate to represent the atmospheric CO profile, it is not the true status. According to the (Rodgers, 2003), the smoothing error should be taken into account when comparing two remote sensing retrievals

$$\sigma_s^2(TC'_{r,N} - TC'_{r,T}) = (\mathbf{A}_N - \mathbf{A}_T)^T \mathbf{PC}_{air}^{dryT} \mathbf{S}_x \mathbf{PC}_{air}^{dry} (\mathbf{A}_N - \mathbf{A}_T), \quad (13)$$

where \mathbf{PC}_{air}^{dry} is the partial column profile of the dry air and \mathbf{S}_x is the a priori covariance estimation of the CO VMR profile in unit of ppb^2 , including systematic and random parts. Since the scaling factor of the NDACC a priori profile is based on the NDACC retrieved total column, and the systematic uncertainty of NDACC XCO data at Izaña, St Denis, Wollongong and Lauder are about 2.0% (see Table 3), it is assumed that the systematic bias for the diagonal values are 2.0%. For Bremen and Ny-Ålesund, the systematic uncertainty might be underestimated. The non-diagonal elements are calculated from the diagonal values $S_{ij} = \sigma_i \sigma_j$ (von Clarmann, 2014). The random part is set as the covariance matrix of the scaled NDACC a priori profiles after smoothing with a correction width of 2.0 km. As an example, the covariance matrix at Bremen is shown in Figure 6. The random covariance is about 10 times larger than the systematic covariance. Table 5 lists the smoothing error when comparing TCCON with NDACC data by using the scaled NDACC a priori profile as the common a priori profile. The systematic smoothing error is within 0.2%, which is relatively small compared to the mean difference between the TCCON and NDACC X_{CO} data (5.6 - 8.5%). The random smoothing error is between 2.0% and 4.2%, which can help to explain the large SD values in the TCCON and NDACC differences. Note that the smoothing error might be underestimated, because the CO profile in the real atmosphere does not always follow the vertical shape of the NDACC a priori profile so that the variability of CO can be larger than what we estimated.

The smoothing errors of the standard TCCON and NDACC CO total column are estimated as

$$\sigma_s^2(TC_{r,T}) = (\mathbf{I} - \mathbf{A}_T)^T \mathbf{PC}_{air}^{dryT} \mathbf{S}_{x,T} \mathbf{PC}_{air}^{dry} (\mathbf{I} - \mathbf{A}_T), \quad (14)$$

$$\sigma_s^2(TC_{r,N}) = (\mathbf{I} - \mathbf{A}_N)^T \mathbf{PC}_{air}^{dryT} \mathbf{S}_{x,N} \mathbf{PC}_{air}^{dry} (\mathbf{I} - \mathbf{A}_N), \quad (15)$$

where the systematic and random covariance matrices $\mathbf{S}_{x,T(N)}$ are calculated from the differences between the scaled NDACC a priori profiles and TCCON (NDACC) original a priori profiles. Table 5 shows that the systematic smoothing error of the TCCON X_{CO} data can reach up to 7.9% (Lauder), which is quite large compared to the difference between TCCON and NDACC X_{CO} measurement. The systematic smoothing error of TCCON data at Southern Hemisphere sites is larger than that at Northern Hemisphere sites. The random smoothing error of TCCON data is in the range between 2.0% and 3.6%, which is

larger than 1.0% estimated in Wunch et al. (2015) by shifting the TCCON a priori CO profile down by 1 km. The systematic smoothing error of NDACC data is in the range between 0.1% and 0.8% and the random smoothing error of NDACC data is about 0.3%. The smoothing error of the TCCON data is much larger than that of the NDACC data, because 1) the TCCON AVK deviates more from 1.0 than the NDACC AVK, and 2) the deviation between the TCCON a priori profile and the true atmosphere seems to be larger than that for NDACC, especially in the Southern Hemisphere.

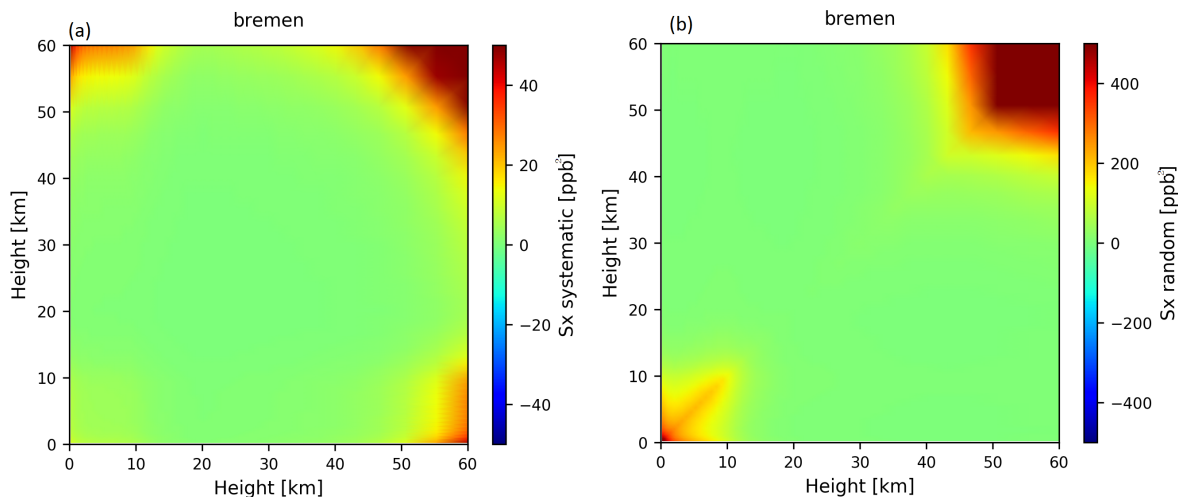


Figure 6. The systematic (a) and random (b) covariance matrices of the common optimal a priori profile (scaled NDACC a priori profiles) at Bremen.

Table 5. The systematic and random smoothing errors of the difference between TCCON and NDACC X_{CO} data (using scaled NDACC a priori profiles as the common a priori profile), standard TCCON X_{CO} data and NDACC X_{CO} data.

Site	Ny-Ålesund	Bremen	Izaña	St Denis	Wollongong	Lauder
σ_s sys/ran [%]	0.1/2.0	0.1/2.4	0.1/2.8	0.2/2.5	0.1/4.2	0.1/2.2
TCCON σ_s sys/ran [%]	3.7/2.0	0.2/2.3	3.0/1.9	5.0/2.1	3.9/3.6	7.9/2.0
NDACC σ_s sys/ran [%]	0.8/0.3	0.3/0.4	0.4/0.1	0.2/0.4	0.1/0.5	0.1/0.2

4.3 Comparison between AirCore and TCCON data

It is found that the difference between the TCCON and NDACC measurements with the common optimal a priori profile is higher than their uncertainties even after taking the smoothing error into account. To investigate the scaling factor (1.0672) of the TCCON X_{CO} data, the AirCore measurements at Sodankylä and Orleans are compared with the TCCON X_{CO} measurements. The AirCore measurements have been performed regularly by the Finnish Meteorological Institute (FMI) and the

University of Groningen (RUG) at Sodankylä (Finland) since September 2013, and by the Laboratoire des Sciences du Climat et de l'Environnement (LSCE) at Orleans (France) since October 2016. Orleans and Sodankylä are operational TCCON sites but there are no NDACC X_{CO} measurements available at these two sites. The AirCore measurement technique uses a balloon to bring a long coiled tube up to the lower or middle stratosphere and samples a vertical profile of air inside the tube during its descent. After its landing, the tube is recovered and the air inside the tube is transferred to a gas analyser to measure the CO mole fraction vertical profile (Karion et al., 2010). As the vertical resolution of the AirCore measurement depends on the molecular diffusion inside the tube, the tubes diameter is kept sufficiently thin (< 1.0 cm) to have a laminar flow at the sampling flow rates (Paul et al., 2016; Membrive et al., 2017). In addition, the AirCore samples were typically analyzed within 4 hours after landing to minimize the influence of molecular diffusion on the vertical resolution of the AirCore profiles. The AirCore measurements covers the vertical range from several hundred meters above the surface to about 20-25 km, and the total uncertainty of the CO measurement is about 2-3 ppb ($\sim 3.0\%$).

To compare the AirCore profiles with the TCCON X_{CO} data, the AirCore profile first needs to be extended to the whole atmosphere. We use the surface in situ measurements (Schmidt et al., 2014; Kilkki et al., 2015) to fill the gap between the surface and the lowest AirCore altitude (several hundred meters above the ground), and the scaled ACE-FTS profile to fill the CO profile above the AirCore altitude to the top of the atmosphere. The ACE-FTS profile is the mean of the all measurements located within $\pm 10^\circ$ latitude band of the FTIR site during 2007-2017. The uncertainties are set as 3.0% for the surface in situ and AirCore measurements, and as 25.0% for the altitude above the AirCore maximum measurement height according to the ACE-FTS data uncertainty (Clerbaux et al., 2008). Second, the “extended” AirCore VMR profile is re-gridded on the TCCON retrieval levels and the partial column profile is calculated based on the surface pressure and NCEP pressure, temperature and water vapor profiles. As an example, Figure 7 shows the “extended” AirCore profile together with the TCCON a priori profile, original AirCore and surface in situ measurements on 15 July 2014 at Sodankylä. Finally, the “extended” AirCore partial column profile is smoothed with TCCON AVK, and the X_{CO} is derived from the smoothed AirCore total column

$$TC_{aircore} = TC_{a,T} + A_T(PC_{aircore} - PC_{a,T}), \quad (16)$$

$$X_{aircore} = TC_{aircore}/TC_{air}^{dry}. \quad (17)$$

The co-located daily mean of the TCCON X_{CO} retrievals is compared with each AirCore measurement. Instead of using 3.0% as the random uncertainty of the TCCON data, the daily SD of the TCCON data is used to represent the random uncertainty of the TCCON data. The scatter plots between the TCCON and AirCore measurements at Orleans and Sodankylä are shown in Figure 8. The TCCON X_{CO} measurements are $6.1 \pm 1.6\%$ and $8.0 \pm 3.2\%$ less than the AirCore measurements at Orleans and Sodankylä respectively. The relative differences between the TCCON and AirCore measurements have no obvious seasonal dependence. This result is consistent with Table 4 showing that the mean NDACC data is 6.8% larger than the TCCON data by using the common optimal a priori profile. Without the scaling factor (or $\alpha = 1.0000$ instead of 1.0672), the mean differences between TCCON and AirCore are $-0.6 \pm 1.6\%$ and $1.3 \pm 3.2\%$ at Orleans and Sodankylä respectively. Further investigations are needed to understand whether the TCCON X_{CO} data are incorrectly scaled at other TCCON sites.

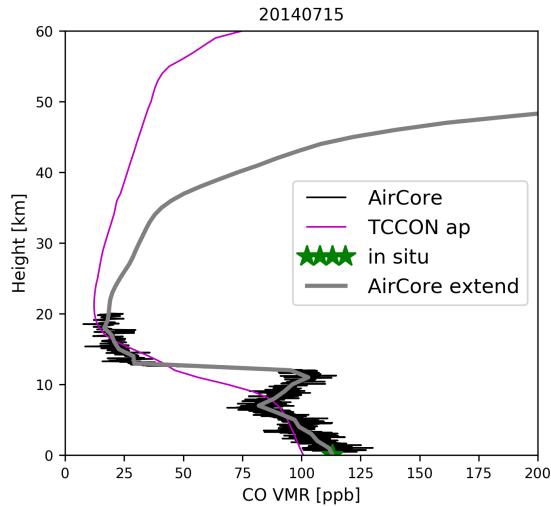


Figure 7. The “extended” AirCore CO profile together with the TCCON a priori profile, original AirCore and surface in situ measurements on 15 July 2014 at Sodankylä.

5 An application example

In this section, we give an example of using the TCCON and NDACC X_{CO} data together to compare against an atmospheric model simulation. The TCCON and NDACC measurements from the six sites are used to compare with the Copernicus Atmosphere Monitoring Service (CAMS) operational (o-suite) reactive gas model re-analysis simulations from March 2015 to December 2018. Because there are no NDACC measurements at St Denis after June 2015, the measurements at Maïdo are used here, which is about 20 km away from St Denis (Zhou et al., 2016). The model uses the chemistry-coupled integrated forecasting system (CIFS) model run with a truncation of T511 which is approximate resolution of 40 km by 40 km and 60 vertical layers (surface to 0.1 hPa). The CAMS o-suite re-analysis CO data have been assimilated with IASI-A, IASI-B and MOPITT satellite measurements (Inness et al., 2015). The model output has a 6-hours temporal resolution. Note that the CAMS o-suite model mainly focuses on the troposphere, and the CO VMR in the stratosphere is underestimated. More information can be found in the CAMS near-real-time system description (<https://confluence.ecmwf.int/display/COPSRV/Global+production+log+files>, last access: 26 April 2019) and the validation report (Wagner et al., 2019).

For each FTIR measurement, the closed CAMS model output in time with space interpolated is selected as one data pair, and an altitude correction is applied to the model output to make the model surface altitude to the same level of the FTIR site (Langerock et al., 2015). The time series of X_{CO} from the FTIR measurements, the CAMS model with and without smoothed with the FTIR data, together with their differences are shown in Figure 9. In general, the model simulates the seasonal variation of X_{CO} very well. However, the model simulation is larger than the FTIR measurements in local winter and smaller than the

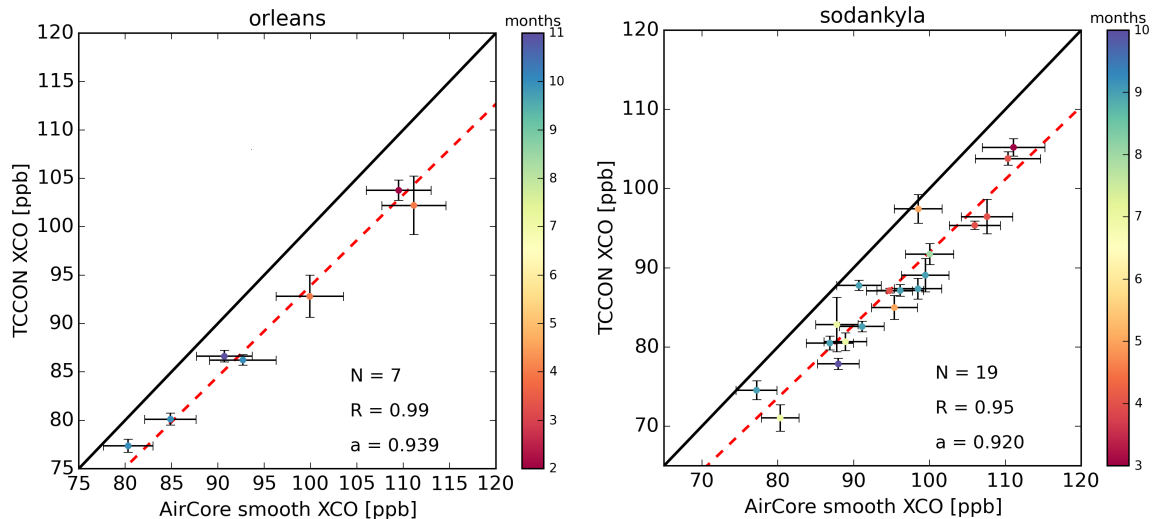


Figure 8. The scatter plots between the TCCON X_{CO} retrievals and the smoothed AirCore X_{CO} measurements at Orleans (left) and Sodankylä (right). The black line is the one-to-one line, and the red dashed line is the linear fitting (forced to cross the zero). The data is colored with the measurement month. The errorbar of the TCCON X_{CO} retrieval is the daily SD, representing the random uncertainty of the TCCON data, while the errorbar of the AirCore data is the total uncertainty for each measurement. N is the number of co-located measurements, R is the correlation coefficient and a is the slope of the fitting line.

FTIR measurements in summer at Ny-Ålesund, indicating an underestimation in the amplitude of the seasonal variation of X_{CO} for the CAMS model at this site. Several high X_{CO} FTIR measurements are not well captured by the CAMS model at Ny-Ålesund and Bremen. Fewer satellite observations improve the CAMS model at higher latitudes due to measurement difficulties, which may cause the poorer performance at these sites. Both TCCON and NDACC measurements show many high X_{CO} values at Wollongong, which are not well simulated in the CAMS model. There is an extremely high value in the CAMS model simulations at Lauder, which is not observed in TCCON and NDACC measurements. High locally impacted values are not expected to be captured by the model due to dilution: both temporally (6 hours compared to minutes) and spatially (40 km² compared to site location).

Table 6 lists the mean and SD of the relative difference between the CAMS model (with and without smoothing) and FTIR measurements. The averaged bias between the TCCON and CAMS smoothed data is 5.2%, while the averaged bias between the NDACC and CAMS smoothed data is -1.2%. The latter bias is due to the underestimation of the stratospheric CO in the CAMS model. The difference between the averaged biases of the CAMS model with TCCON and NDACC data is 6.4%, which is consistent with the result obtained when comparing TCCON and NDACC X_{CO} data using the scaled NDACC a priori profile as the common a priori profile (see Table 4). According to the AirCore measurements in Section 4.3, the bias of 5.2% between the TCCON and CAMS smoothed data is mainly due to the scaling factor of the TCCON X_{CO} measurements. In addition, Table 6 shows that the changing of the model X_{CO} data after smoothing with TCCON data ranges from 2.1% (Bremen) to 6.1%

(Lauder), which is much larger than that after smoothing with NDACC data of 0.3 - 2.4%. It is confirmed that the smoothing error of TCCON X_{CO} data is much larger than that of NDACC X_{CO} data, and the smoothing error must be taken into account when using FTIR X_{CO} data.

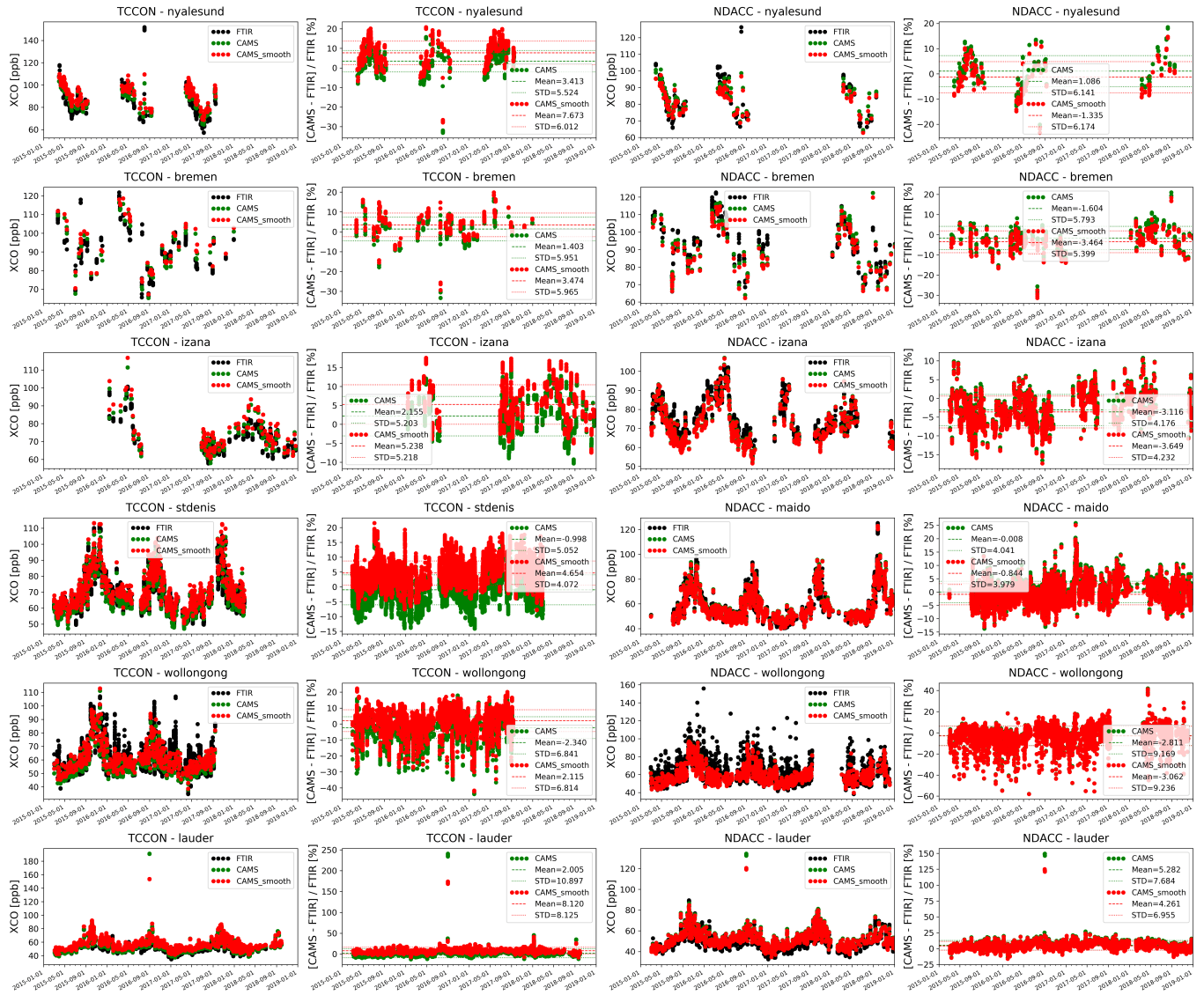


Figure 9. The time series of X_{CO} from the TCCON measurements, the CAMS model and the CAMS model smoothed with TCCON data at six sites (first column) and their relative differences (second column). The time series of X_{CO} from the NDACC measurements, the CAMS model and the CAMS model smoothed with NDACC data at six sites (third column) and their relative differences (last column).

Table 6. The mean and SD of the relative difference between the CAMS and FTIR (TCCON and NDACC) X_{CO} data, with and without smoothing. St Denis*: TCCON data is from St Denis site, while NDACC data is from Maïdo site.

(CAMS-FTIR)/FTIR [%]	TCCON	TCCON smooth	NDACC	NDACC smooth
Ny-Ålesund	3.4±5.5	7.6±6.0	1.1±6.1	-1.3±6.1
Bremen	1.4±6.0	3.5±6.0	-1.6±5.8	-3.5±5.4
Izaña	2.1±5.2	5.2±5.2	-3.1±4.2	-3.6±4.2
St Denis*	-1.0±5.1	4.7±4.1	-0.0±4.0	-0.8±4.0
Wollongong	-2.3±6.8	2.1±6.8	-2.8±9.2	-3.1±9.2
Lauder	2.0±10.9	8.1±8.1	5.3±7.7	4.3±7.0

6 Conclusions

In this study, the difference between the TCCON and NDACC X_{CO} data products during the period 2007-2017 has been studied at six sites (Ny-Ålesund, Bremen, Izaña, St Denis, Wollongong and Lauder) where co-located NDACC and TCCON FTIR observations are carried out.

- 5 When doing a straightforward comparison between both X_{CO} data products, it is found that for the Northern Hemisphere sites the TCCON X_{CO} values are about 5.5% smaller than the NDACC X_{CO} values, and the absolute bias between the NDACC and TCCON data are within 2% at the Southern Hemisphere sites. To understand these inter-hemispheric differences in the biases, we have looked into more detail in the characteristics of both products, in particular their averaging kernels and dependence on the a priori profiles used in the retrievals. Taking into account these differences in the comparisons, by adjusting
- 10 the products towards a common optimal a priori profile, it is found that the biases between the adjusted TCCON and NDACC X_{CO} data products are almost constant (5.6 - 8.6%) with a mean value of 6.8%; for the common optimal a priori profile we have chosen the NDACC a priori profiles scaled with the ratios of the retrieved to the a priori columns.

The first conclusion therefore is that the apparent inter-hemispheric difference in the bias disappears when accounting correctly for the smoothing errors. To confirm this first finding we have estimated the systematic and random smoothing errors of

15 the TCCON and NDACC X_{CO} data according to the optimal estimation method (Rodgers, 2000): the TCCON X_{CO} systematic smoothing errors vary in the range between 0.2% (Bremen) and 7.9% (Lauder), and their random smoothing errors lie in the range between 2.0% and 3.6%, which is larger than the random uncertainty of 1.0% estimated in Wunch et al. (2015). Also the TCCON X_{CO} systematic and random smoothing errors are larger than the NDACC X_{CO} systematic and random smoothing errors that are in the range between 0.1% and 0.8% for the systematic ones and of order 0.3% for the random ones, and

20 they are larger in the Southern than in the Northern hemisphere. This is because 1) the TCCON AVK deviates more from 1.0 than the NDACC AVK, and 2) the deviation between the TCCON a priori profile and the true profile seems to be larger than that for NDACC, especially in the Southern Hemisphere. This finding also demonstrates the importance of accounting for the smoothing errors when comparing FTIR X_{CO} data, and particularly TCCON X_{CO} data, with satellite measurements or model simulations. This has not always been done in recent satellite validation studies (Borsdorff et al., 2016, 2018; Hochstaff et al.,

2018). As a consequence, the biases reported in these papers are not relevant because they fall in the systematic uncertainty, especially in the Southern Hemisphere.

Our second conclusion is that the remaining 6.8% bias between the TCCON and NDACC X_{CO} data (when using the common optimal a priori profile) originates in the scaling correction that has been applied to the standard TCCON data. To demonstrate this second finding we have compared AirCore in situ profile measurements with the standard TCCON X_{CO} data. It is found that the TCCON X_{CO} measurements are $6.1 \pm 1.6\%$ and $8.0 \pm 3.2\%$ smaller than the AirCore measurements at Orleans and Sodankylä, respectively, which is consistent with the bias found between the TCCON and NDACC X_{CO} measurements. Eliminating the scaling correction (setting $\alpha = 1.0000$ instead of 1.0672), the differences between the TCCON and AirCore measurements become $-0.6 \pm 1.6\%$ and $1.3 \pm 3.2\%$ at Orleans and Sodankylä, respectively. A similar confirmation is found when comparing the TCCON X_{CO} data to CAMS assimilation analyses. Further investigations should therefore be carried out in the TCCON community to study the CO scaling factor based on comparisons with in situ CO profile observations (e.g. calibrated aircraft or AirCore measurements) at additional TCCON sites.

Data availability. The TCCON GGG2014 data are publicly available through the TCCON database (<https://tccodata.org/>). The details of the TCCON data for each site, please refer to Notholt et al. (2017, 2014); Blumenstock et al. (2014); De Mazière et al. (2014); Griffith et al. (2014); Sherlock et al. (2014); Warneke et al. (2014); Kivi et al. (2014); Kivi and Heikkinen (2016). The NDACC data are publicly available from the NDACC website (<http://www.ndacc.org>).

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This study is supported by the EU-funded C3S_311a_Lot3 project. The TCCON site at Reunion Island is operated by the Royal Belgian Institute for Space Aeronomy with financial support in 2014, 2015, and 2016, 2017 under the EU project ICOS-Inwire and the ministerial decree for ICOS (FR/35/IC2) and local activities supported by LACy/UMR8105 - Université de La Réunion. The NDACC and TCCON stations Ny-Ålesund, Bremen, Izaña have been supported by the German Bundesministerium für Wirtschaft und Energie (BMWi) via DLR under grants 50EE1711A-B. The Lauder FTIR measurements are core funded by NIWA from New Zealand's Ministry of Business, Innovation and Employment through the Strategic Science Investment Fund. We thank the HIPPO team for making the aircraft measurements available <http://hippo.ucar.edu/>.

Author contributions. MZ, CV and MDM designed the study. MZ wrote the paper and produced the main analysis and results with significant input from BL. HC and MR, RK provide the AirCore measurements. MKS, CH, JMM, PH, DS, DFP, NJ, VAV, OEG, MS, MP, TW provided and analyzed the TCCON and NDACC measurements. All authors read and provided comments on the paper.

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