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Retrieval of xCO$_2$ from ground-based mid-infrared (NDACC) solar absorption spectra and comparison to TCCON

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Abstract. High-resolution solar absorption spectra, taken within the Network for the Detection of Atmospheric Composition Change Infrared Working Group (NDACC-IRWG) in the mid-infrared spectral region, are used to infer partial or total column abundances of many gases. In this paper we present the retrieval of a column-averaged mole fraction of carbon dioxide from NDACC-IRWG spectra taken with a Fourier transform infrared (FTIR) spectrometer at the site in Ny-Ålesund, Spitsbergen. The retrieved time series is compared to colocated standard TCCON (Total Carbon Column Observing Network) measurements of column-averaged dry-air mole fractions of CO$_2$ (denoted by xCO$_2$). Comparing the NDACC and TCCON retrievals, we find that the sensitivity of the NDACC retrieval is lower in the troposphere (by a factor of 2) and higher in the stratosphere, compared to TCCON. Thus, the NDACC retrieval is less sensitive to tropospheric changes (e.g., the seasonal cycle) in the column average.

1 Introduction

The continued measurement of atmospheric trace gases, especially in the context of climate change, has given valuable insight on their sources and sinks (Ciais et al., 2013). In situ measurements offer high accuracy and repeatability but are spatially highly constrained and influenced by local perturbations. Remote-sensing instruments that sample the total column are less influenced by boundary layer processes and can provide complementary information to in situ measurements because they sample the total atmospheric column.

A worldwide network of Fourier transform infrared (FTIR) spectrometers acquires solar absorption spectra from which total columns of a number of gases are retrieved. The Infrared Working Group within the Network for the Detection of Atmospheric Composition Change (NDACC-IRWG) provides time series of mid-infrared (MIR) solar absorption spectra at various sites dating back to 1978 (Kitt Peak Observatory). Since its inception in 2004, the Total Carbon Column Observing Network (TCCON) has created a high-quality database of near-infrared (NIR) solar absorption spectra. Both networks have been used to deduce total column amounts of various atmospheric constituents, the main focus being centered around O$_3$ chemistry (NDACC; see, e.g., Notholt et al., 1995) and the carbon cycle (TCCON; see, e.g., Wunch et al., 2011a).

TCCON retrievals of the dry-air mole fraction of CO$_2$ employ a method of taking the ratio of CO$_2$ from the 6300 cm$^{-1}$ band to O$_2$ which is measured simultaneously from the 7885 cm$^{-1}$ band (Yang et al., 2002). This approach compensates for common systematic errors like light path variations and pointing errors. Additionally, the selected spectral windows in the NIR have few interfering gases, allowing for high-quality spectral fits. This results in a network-wide accuracy of the TCCON data of around 0.2% for xCO$_2$.

However, near-infrared TCCON measurements have only been available since 2004, with many sites starting operation...
more recently. An extension of the retrieval capabilities to the mid-infrared NDACC spectra would not only expand the total column products in time but would also expand the spatial coverage. Two major challenges have to be faced on the way to a NDACC xCO₂ product: in contrast to the NIR, there are no O₂ absorption lines present in the MIR spectra; therefore there is no proxy for the dry-air column to which to take a ratio. Additionally the presence of multiple interfering gases hinders the use of broad spectral windows.

This study was undertaken within the framework of ESA’s Greenhouse Gas Climate Change Initiative (Buchwitz et al., 2013). Its aim is to assess whether MIR FTIR spectra could be used to derive a total column CO₂ mole fraction. Sussmann et al. (2013) performed a similar study on the inter-comparison of methane from MIR and NIR retrievals. They used a Tikhonov L1 regularized profile retrieval scheme with the standard NDACC retrieval package SFIT2 for the MIR and the standard TCCON retrieval (GFIT) for the NIR spectra. They conclude that the information obtainable from the retrieval is significantly different between MIR and NIR retrievals, which leads to different seasonal variations.

Recently, Barthlott et al. (2015) used MIR xCO₂ retrievals in combination with a simple model to investigate the long-term consistency of NDACC measurements. They conclude that the low sensitivity of the MIR averaging kernels in the lower troposphere is sufficient to perform comparisons to the model on timescales longer than monthly, and therefore xCO₂ retrievals in the MIR are appropriate for monitoring long-term instrument stability. However, the annual increase of xCO₂ is already well understood; in order to gain new insights into carbon cycle processes, information on shorter timescales is needed to assess and complement model simulations. Additionally, the analysis of systematic differences in Barthlott et al. (2015) is performed using monthly means, and the dampening of the seasonal cycle amplitude is proposed to be addressed using a dampening factor derived from the shape of the site’s pressure profile. Although this is adequate for the paper’s goal of monitoring instrument stability, a complete analysis of the different sensitivities of the MIR and NIR retrievals is necessary for the retrieval of information relevant to carbon cycle research.

In this paper we describe the retrieval of the column average dry-air mole fraction of CO₂ (denoted xCO₂) from MIR spectra and discuss the differing retrieval sensitivities from the NIR and the dependency of the retrieval on the a priori information using the same retrieval code on both data sets. We conclude that it is possible to retrieve xCO₂ from NDACC spectra. The secular increase of xCO₂ is well captured. Information on the seasonal cycle, however, is mainly introduced by the a priori. We show that this is caused by the different averaging kernels, which show that the NDACC xCO₂ retrievals are most sensitive to the stratospheric CO₂. We investigate the averaging kernel smoothing correction and show that it is not able to compensate for the different sensitivities. This would be a necessary step in proving that a direct substitution of a NIR xCO₂ product or usage of the MIR product as a complementary source of information is possible.

In the second section we describe the microwindows used in the retrieval and then continue with a description of the retrieval methods for the MIR and briefly for the NIR. In the third and fourth sections we investigate the influence of both the a priori and averaging kernels on the retrieval in comparison to TCCON. We conclude with a summary of the results.

2 Methods

2.1 Measurement site

For this study we chose the station in Ny-Ålesund (78.92° N, 11.92° E) for its long time series and the large seasonal cycle of xCO₂. The instrument used is a Bruker IFS 120HR FTIR spectrometer. Measurements of MIR spectra have been taken since 1992. Since 2005, NIR spectra have also been measured taken within TCCON. Measurements are taken under cloud-free conditions alternately in the MIR and NIR. Due to its high latitude, no solar absorption measurements can be taken in Ny-Ålesund during the polar night.

2.2 CO₂ microwindows in the mid-infrared

In the mid-infrared region between 2000 and 4000 cm⁻¹ in addition to CO₂, other, interfering gases have strong absorption lines. Some spectral regions are dominated by the interference gases, while in others CO₂ is the most prominent feature. This makes it impractical to select whole absorption bands for the retrieval, such as is done in the NIR by the TCCON. The high-resolution NDACC spectra allow the selection of microwindows that contain only single lines. The contribution of interfering gases to the single line shapes varies for different microwindows.

Multiple microwindows are fitted in three spectral regions: four microwindows at around 2624 cm⁻¹, two microwindows at 3160 cm⁻¹ and two around 3330 cm⁻¹. The corresponding line parameters are taken from the GGG line list, which is based on the HITRAN (high-resolution transmission molecular absorption) database (Rothman et al., 2005, 2009). These are shown in Table 1. Each of the spectral regions referred to above corresponds to a different optical filter or detector setting, and they are measured sequentially at acquisition. The retrieval is performed separately for each spectral interval and microwindow. Following this approach, we obtain eight different retrievals of xCO₂, one for each microwindow.

Retrievals from microwindows within the same spectral interval were averaged and used to derive daily averages to correlate with the daily-averaged TCCON retrievals. This approach circumvents the temporal coincidence problem that arises due to the different acquisition procedures described in Sect. 2.1.
Table 1. The mid-IR microwindows used in this study. Each window is characterized by the center wavenumber and its width. Additionally, the corresponding molecular transition is given. Data taken from Rothman et al. (2005, 2009). Example spectral microwindows are shown in Fig. 1.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Center [cm⁻¹]</th>
<th>Width [cm⁻¹]</th>
<th>Quantum transition</th>
<th>Line intensity [cm⁻¹ (molecule cm⁻²)]</th>
<th>Lower state energy [cm⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>16O12C18O</td>
<td>2620.83</td>
<td>0.55</td>
<td>20002 → 00001</td>
<td>3.270 × 10⁻²⁵</td>
<td>26.5087</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>2626.63</td>
<td>0.45</td>
<td>20002 → 00001</td>
<td>4.309 × 10⁻²⁵</td>
<td>100.1374</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>2627.35</td>
<td>0.50</td>
<td>20002 → 00001</td>
<td>4.291 × 10⁻²⁵</td>
<td>112.6533</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>2629.61</td>
<td>0.67</td>
<td>20002 → 00001</td>
<td>4.076 × 10⁻²⁵</td>
<td>154.6166</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>3160.22</td>
<td>0.16</td>
<td>21103 → 00001</td>
<td>3.140 × 10⁻²⁵</td>
<td>316.7698</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>3161.70</td>
<td>0.20</td>
<td>21103 → 00001</td>
<td>3.030 × 10⁻²⁵</td>
<td>273.868</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>3315.78</td>
<td>0.55</td>
<td>21102 → 00001</td>
<td>1.726 × 10⁻²⁴</td>
<td>362.7882</td>
</tr>
<tr>
<td>16O12C18O</td>
<td>3344.81</td>
<td>0.26</td>
<td>21102 → 00001</td>
<td>7.747 × 10⁻²⁵</td>
<td>16.389</td>
</tr>
</tbody>
</table>

Figure 1. An example fit of the measured (black) with the calculated (green) spectrum and their residuals (blue) for the various microwindows used. Note the different signal-to-noise ratio due to different instrument setup (see Sect. 2.2).

2.3 xCO₂ retrieval

The MIR xCO₂ retrieval has been performed with the profile scaling algorithm GFiT (version 4.8.6 in GGG2012), which is also used for the TCCON xCO₂ retrievals. In GFiT a priori profiles of the target gas; gases of interest; and pressure, temperature and H₂O are convolved with an instrument line-shape model to simulate a spectrum. The gas profiles are scaled and other spectral parameters adjusted to provide the best fit to the measured spectrum by minimizing the residual. From these scaled profiles, the amount of the target gas is retrieved. An example fit is shown in Fig. 1.

The standard TCCON retrieval uses O₂ retrieved from the same spectra as the target gases in the band at 7885 cm⁻¹ to estimate the total dry-air column. The retrieved CO₂ column is then divided by this retrieved O₂ column to derive the column average dry-air mole fraction. The method is explained in further detail in Wunch et al. (2011a).

The same software version was used to analyze the MIR spectra. To benefit from the abovementioned ratio approach, a MIR active species with a well-known atmospheric abundance would be needed. Only N₂ has some weak absorption lines in the target region: its multipolar 1 → 0 transition. The suitability of these lines for atmospheric retrieval was previously investigated by Goldman et al. (2007). Due to the very low intensity, the retrieved N₂ mole fractions introduced more noise than was compensated by use of the ratio, meaning that N₂ was not able to be used as an internal standard.

Instead, mole fractions from the MIR spectra are calculated by normalization by the dry-air column abundance inferred from an independent measurement of surface pressure. The vertical column of H₂O was also retrieved from a dedicated H₂O microwindow in the vicinity of the respective CO₂ microwindow. Total column CO₂ was then calculated as described in Appendix A in Wunch et al. (2011b) by applying

\[
\text{xCO}_2 = \frac{V_{\text{CO}_2}}{P_{\text{air}} \langle g_{\text{air}} \rangle} \frac{m_{\text{dry}}}{m_{\text{H}_2\text{O}}} \frac{V_{\text{H}_2\text{O}} m_{\text{H}_2\text{O}}}{V_{\text{H}_2\text{O}} m_{\text{H}_2\text{O}}}.
\]

Here, \(m_{\text{dry}}\) and \(m_{\text{H}_2\text{O}}\) denote the molecular masses of dry air and water, respectively; \(P_{\text{air}}\) the surface pressure; and \(\langle g_{\text{air}} \rangle\) the column-averaged gravitational acceleration. \(V_{\text{CO}_2}\) and \(V_{\text{H}_2\text{O}}\) are the retrieved vertical column amounts from the GFiT output.

The retrieved dry-air mole fractions are then quality-controlled by removing data points with high solar zenith angles (> 80°) and/or with a high relative retrieval error (> 20%). Table 2 shows uncertainty estimates for the resulting time series, based on the standard deviation of the daily means. The mean of the standard deviations for the three microwindow regions is compared to the standard TCCON results. Additionally the standard error is shown in order to better reflect the uncertainty in the daily mean, which is also influenced by the number of measurements on each day.
Table 2. Overview over the estimated errors from the different retrievals. Given are both the mean standard deviation of the daily mean and the mean standard error for $N$ measurements, in ppm.

<table>
<thead>
<tr>
<th></th>
<th>TCCON 2600 cm$^{-1}$</th>
<th>TCCON 3100 cm$^{-1}$</th>
<th>TCCON 3300 cm$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma$</td>
<td>1.13</td>
<td>2.06</td>
<td>3.21</td>
</tr>
<tr>
<td>$\sigma / \sqrt{N}$</td>
<td>0.23</td>
<td>0.96</td>
<td>1.67</td>
</tr>
</tbody>
</table>

Only days with two or more measurements have been used. This approach results in an error estimate ($1\sigma$) of 1.13 ppm for the Ny-Ålesund TCCON standard product ($\approx 0.3\%$) and 2.06 ppm (0.5\%) for the 2600 cm$^{-1}$ microwindow (see Table 2).

As a basis for the analysis, daily means of in total 335 days (TCCON) and 223 days (NDACC) between March 2005 and May 2013 were used. The average number of measurements per day was about 30 for TCCON and 5 for the NDACC 2000 cm$^{-1}$ region.

3 Influence of a priori information

Following Rodgers (2000) and Rodgers and Connor (2003), the retrieval of the total column abundance $\hat{x}$ (for a true quantity $x$) as described in Sect. 2.3 depends on both the choice of the a priori profile $x_a$ and the measurements averaging kernel $A$.

Thus, Eq. (3) in Rodgers and Connor (2003) gives

$$\hat{x} - x_a = A(x - x_a) + \epsilon_x,$$

where $\epsilon_x$ is the random and systematic error term. If the measurement information content does not fully constrain the retrieval, the result will be influenced by the a priori. Within TCCON a latitude- and time-dependent empirical model is used to simulate an $x$CO$_2$ profile that is already quite close to the true atmospheric profile. There is one TCCON a priori profile per day calculated for local noon. These are shown in Fig. 2.

Usage of this modeled a priori profile in the MIR retrieval can give an overly favorable impression of the information content of the MIR retrieval. Even if there is no additional information to be retrieved from the spectra, a reasonably good agreement between the NIR and MIR retrievals is still to be expected, because the smoothing error term tends to zero when the prior is approximately equal to the true atmospheric state (for a detailed explanation see Sect. 4).

To investigate this, $x$CO$_2$ has been retrieved from both NIR and MIR spectra, once using the standard, time-dependent TCCON a priori, and once using a fixed, constant a priori profile of 379 ppm throughout the atmosphere. Apart from the different a priori, the same retrieval and post-processing parameters have been used as in the original retrievals with the TCCON a priori.

The comparison of the NIR (TCCON) time series with modeled and fixed a prioris shows a clear pattern. This corresponds to an overestimation of the seasonal cycle amplitude by the retrieval using a fixed a priori compared to the standard a priori. At seasonal maximum the difference is negative, and at the seasonal minimum positive. The same pattern can be deduced from the slope in the correlation plot in Fig. 3. This can be attributed to the usage of a fixed a priori in conjunction with the solar-zenith-angle-dependent averaging kernel (see Fig. 4). The solar zenith angles are generally larger in spring and autumn than in summer. The TCCON averaging kernels show an increased sensitivity in the troposphere. This coincides with the seasonal maximum of $x$CO$_2$ in spring and with the seasonal minimum in autumn. In these cases the TCCON averaging kernel diverges from being close to 1, and subsequently the smoothing error contributions increase if the prior differs from the true atmospheric state. Thus the residual pattern in Fig. 3 originates from the shape of the averaging kernels.

In the case of the MIR NDACC spectra, the results of the retrieval with the TCCON a priori in comparison to that with a fixed a priori can be seen in Fig. 5. Both time series (the NDACC retrieval with the TCCON a priori and that with the fixed a priori) capture the secular trend, and no residual trend is visible. The seasonal cycle amplitude, however, is damped in the NDACC retrieval (shown here are the results of the 2600 cm$^{-1}$ microwindow; other regions show comparable results). A clear seasonal structure can be seen in the residual.

In order to quantify the effect that the different a priori information has on the retrievals, a simple function (see Eq. 3) has been fitted to each time series (see Fig. 6):

$$x$CO$_2$(t) = a + b \cdot t + A \cdot \sin(2\pi \cdot t),$$

where $a$ is an offset at the beginning of the partial time series (1 January 2009), $b$ is the linear trend per year and $A$ is the amplitude of the seasonal cycle. The fit parameters are pre-
Figure 3. (a) $x$CO$_2$ from the TCCON standard window for retrievals using the TCCON (upper panel) and fixed (center panel) a priori and their difference (lower panel). (b) The corresponding correlation plot of the detrended $x$CO$_2$ time series of the TCCON standard window, with modeled and fixed a priori.

Table 3. Fit parameters corresponding to the fits of the time series in Fig. 6 and in Eq. (3). Units are xCO$_2$ in ppm as a function of time in decimal years starting 1 January 2009. Uncertainty estimates are $1\sigma$ fitting errors.

<table>
<thead>
<tr>
<th>Retrieval</th>
<th>$a$ [ppm]</th>
<th>$b$ [ppm yr$^{-1}$]</th>
<th>$A$ [ppm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>TCCON</td>
<td>383.27 ± 0.20</td>
<td>2.23 ± 0.09</td>
<td>5.73 ± 0.14</td>
</tr>
<tr>
<td>TCCON fixed</td>
<td>383.55 ± 0.19</td>
<td>2.21 ± 0.08</td>
<td>6.55 ± 0.14</td>
</tr>
<tr>
<td>NDACC</td>
<td>381.93 ± 0.64</td>
<td>2.41 ± 0.24</td>
<td>5.60 ± 0.49</td>
</tr>
<tr>
<td>NDACC fixed</td>
<td>381.08 ± 0.67</td>
<td>2.44 ± 0.28</td>
<td>2.64 ± 0.50</td>
</tr>
</tbody>
</table>

sent in Table 3. The comparison of the fitted amplitudes for the different retrievals allows for an estimate of the magnitude of the dampened seasonal cycle amplitude.

The results show a slight increase from the case of the standard TCCON retrieval to the TCCON retrieval with a fixed a priori by a factor of 1.14 (as was discussed earlier and can be seen in the correlation plot in Fig. 3). However, the fixed a priori NDACC retrieval shows a decrease in the seasonal cycle amplitude. The amplitude of the NDACC retrieval with modeled a priori is found to be 2.12 times larger than the amplitude of the fixed a priori NDACC retrieval.

Similar to the approach used above for the TCCON case, we now investigate whether this is due to the differences in the averaging kernel, i.e., the sensitivity of the retrievals to different height levels. The tropospheric (or stratospheric) contribution of a retrieval can be estimated by integrating the averaging kernels for a retrieval (normalized on a pressure grid) over the contributing pressure column. Thus, the averaging kernels have been integrated in two parts, separated at the tropopause height at 300 hPa (see Fig. 7).

As can be seen in Fig. 4, in contrast to the NDACC xCO$_2$ kernels the TCCON averaging kernels strongly vary with the solar zenith angle. This leads to a zenith-angle-dependent ratio between the stratospheric and tropospheric contribution of the averaging kernels.

For the presented time series of NDACC xCO$_2$ from the 2600 cm$^{-1}$ microwindow the average solar zenith angle is 69°. For this angle, a ratio of 2.05 of the tropospheric averaging kernel contribution of TCCON to NDACC xCO$_2$ can be calculated. Values of 1.80 and 2.18 are calculated for solar zenith angles of 40 and 80°, respectively. Assuming that variability in CO$_2$ occurs solely in the troposphere (i.e., that the seasonal variation in the stratosphere is negligible), a change in tropospheric CO$_2$ would result in a signal that is retrieved as being 2 times smaller than that retrieved from TCCON. This is in good agreement with the tropospheric contribution calculated from the time series’ fits with a fixed a priori described above (2.12).

4 Averaging kernel smoothing error estimation

In the last section, the influence of a varying a priori profile on the retrieval results was investigated. In the following the effect the different shape of the averaging kernels (given a common a priori) has on the retrieved xCO$_2$ is addressed. The differently shaped averaging kernels (see Fig. 4) must be taken into account in a direct comparison of the NDACC and TCCON xCO$_2$ products (Rodgers and Connor, 2003).

The general difference in the averaging kernel shape originates from the (compared to the NIR) narrow absorption lines in the MIR. Thus the NDACC xCO$_2$ averaging kernels show a higher sensitivity at lower atmospheric pressures.

We can use a common prior to quantify the differences between the two retrievals we expect to find for a given true atmospheric state. This is then an estimate of the averaging kernel smoothing error. Following the derivation in Sect. 4.3 in Rodgers and Connor (2003) and in Appendix A of Wunch et al. (2011b), two total column retrievals of a remote-sounding instrument can be compared by choosing a common a priori $x$ and a true atmospheric CO$_2$ profile $\tilde{x}$. The difference of two retrieved total column amounts con-
Figure 4. Left: the total column averaging kernels from the TCCON retrievals. The official averaging kernels from the Lamont site, linearly interpolated to obtain intermediate profiles. Note the minimal possible solar zenith angle in Ny-Ålesund is 55°. Right: the total column averaging kernels from the 2626 cm$^{-1}$ microwindow as an example NDACC averaging kernel. Note the high sensitivity in the stratosphere and low sensitivity in the troposphere in comparison to TCCON.

Figure 5. (a) $\text{xCO}_2$ from the 2600 cm$^{-1}$ NDACC spectra for retrievals using a modeled (upper panel) and fixed (center panel) a priori and their differences (lower panel). No residual secular trend is evident in the residuum. A clear seasonal pattern is visible, and thus a big part of the seasonal cycle is damped, when using a fixed a priori. (b) The corresponding correlation plot of the de-trended $\text{xCO}_2$ time series of TCCON vs. the NDACC retrievals with modeled and fixed a priori.

Figure 6. Fits to the different retrievals. Top two panels: standard TCCON retrieval and TCCON retrieval with fixed a priori. Bottom two panels: NDACC with TCCON and fixed a priori. The fit parameters corresponding to Eq. (3) are presented in Table 3. For clarity, only a subset of the data from 2009 to 2013 is shown.

distributed by the different averaging kernels is then

$$c_{\text{corr}} = \frac{1}{p_0} \sum_j \Delta p_j \left( a_{\text{mir}} - a_{\text{nir}} \right)_j \left( x - x_a \right)_j,$$

with the surface pressure represented by $p_0$, the column averaging kernels by $a$, true profile by $x$ and the common a priori by $x_a$. As the same a priori information is used in both the NDACC and TCCON retrievals, the common a priori $x_a$ is simply the TCCON a priori. The single elements are summed over the profile’s pressure levels $j$, with $\Delta p_j$ being the pressure difference between two levels. The NIR averaging kernels are mostly solar zenith angle dependent, and the Lamont averaging kernels interpolated to the respective zenith angles were used (as suggested by the TCCON database guidelines).
Furthermore, this approach necessitates the knowledge of a true atmospheric profile of the target species (x). As there are no independent measurements of the true atmospheric CO$_2$ profile with sufficient spatial and temporal coverage available, we use a model profile for this estimation.

As a true profile we chose a simulation from NOAA’s CarbonTracker (CT2013) project (CarbonTracker, 2013; Peters et al., 2007), linearly interpolated to the site location and to the atmospheric pressure levels used in our retrieval. This allows for an estimation of the influence of the averaging kernel smoothing contribution between the two retrievals.

In Fig. 8, the averaging kernel correction for the Ny-Ålesund time series has been calculated (as shown above) for all data points. Afterwards the data from the different microwindows around 2600 cm$^{-1}$ have been combined, and daily averages computed.

The residual in Fig. 8 shows a varying influence of the correction that is associated with the solar zenith angle of the measurements. The magnitude of the correction is solar zenith angle dependent and generally between 1 and −1 ppm. Results from the other microwindow regions show comparable behavior.

5 Conclusions

The presented approach for the retrieval of xCO$_2$ from NDACC MIR microwindows can be used to retrieve information about the total column mole fraction of CO$_2$. However, further analysis shows a strong sensitivity to the chosen a priori, as shown previously by Barthlott et al. (2015).

The information content of NDACC mid-infrared spectra can constrain the retrieval to robustly reproduce a secular increase of xCO$_2$. We show that the tropospheric signal (e.g., the seasonal cycle amplitude) is damped in comparison to TCCON. This can be traced back to the shape of the mid-infrared averaging kernels of the NDACC retrieval, which show little sensitivity in the troposphere and high sensitivity in the stratosphere. The NDACC xCO$_2$ product can be used for the secular increase, but this does not provide additional information above what is already provided by in situ measurements. The inability to better resolve the seasonal cycle also limits the use of this product for satellite validation. Consequently, we conclude that these retrievals offer limited value for use in carbon cycle studies.

A direct comparison of the TCCON and NDACC products remains challenging, due to their complementary sensitivities. Furthermore, the specific sensitivity of the NDACC averaging kernels suggests the possibility of investigating stratospheric changes. We anticipate that a full CO$_2$ profile retrieval, rather than the profile scaling approach used here, will be required to make best use of NDACC mid-infrared spectra in carbon cycle research.

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