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# Source brightness fluctuation correction of solar absorption Fourier Transform mid infrared spectra

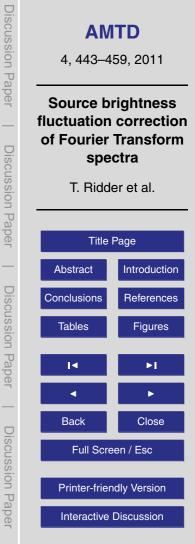
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#### Abstract

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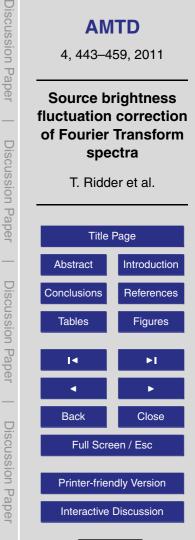
Solar absorption Fourier Transform infrared spectrometry is considered a precise and accurate method for the observation of trace gases in the atmosphere. The precision and accuracy of such measurements are dependent on the stability of the light source.

<sup>5</sup> Fluctuations in the source brightness reduce the precision and accuracy of the trace gas concentrations, but cannot always be avoided. Thus, a strong effort is made within the community to reduce the impact of source brightness fluctuations by applying a correction on the spectra following the measurements. So far, it could be shown that the precision and accuracy of CO<sub>2</sub> total column concentrations could be improved by ap-<sup>10</sup> plying a source brightness fluctuation correction to spectra in the near infrared spectral region.

The analysis of trace gas concentrations obtained from spectra in the mid infrared spectral region is fundamental. However, spectra below 2000 cm<sup>-1</sup> are generally considered uncorrectable, if they are measured with a MCT detector. Such measurements introduce an unknown offset to MCT interferograms, which prevents a source brightness fluctuation correction.

Here, we show a method of source brightness fluctuation correction, which can be applied on spectra in the whole infrared spectral region including spectra measured with a MCT detector. We present a solution to remove the unknown offset in MCT interferograms allowing MCT spectra for an application of source brightness fluctuation correction. This gives an improvement in the quality of MCT spectra and we demonstrate an improvement in the retrieval of  $O_3$  profiles and total column concentrations.

For a comparison with previous studies, we apply our source brightness fluctuation correction method on spectra in the near infrared spectral region and show an improve-<sup>25</sup> ment in the retrieval of CO<sub>2</sub> total column concentrations.





### 1 Introduction

Ground-based solar absorption Fourier Transform infrared (FTIR) spectrometry (Zander et al., 1983; Goldman et al., 1988; Rinsland et al., 1991) has been established as an accurate and precise method for the detection of trace gases in the atmosphere

<sup>5</sup> (Notholt et al., 2003; Velazco et al., 2005). FTIR spectrometry is used by the Network for the Detection of Atmospheric Composition Change (NDACC, 1991) and the Total Carbon Column Observing Network (TCCON, 2005) for the worldwide observation of trace gases.

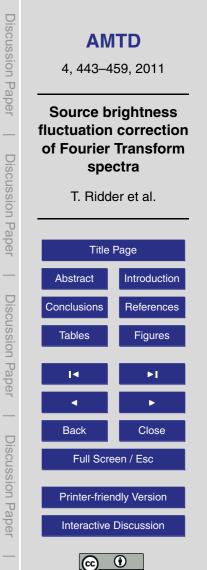
The accuracy and precision of trace gas concentrations retrieved from FTIR spectra
depend on the stability of the light source during the measurement (Beer, 1992; Notholt et al., 1997), since intensity fluctuations can distort the fractional line depth in FTIR spectra (Keppel-Aleks et al., 2007). Variations in the source brightness caused e.g. by clouds decrease the accuracy and precision, but cannot always be avoided. Thus, one aim of the NDACC and TCCON networks is to reduce the impact of source brightness
fluctuations (SBFs) on FTIR spectra by applying a correction to the spectra following the measurements.

The idea of SBF correction was primarily published by Brault (1985) and was recently picked up by Keppel-Aleks et al. (2007). In general, a SBF correction can be applied on Fourier Transform DC interferograms. The measured raw interferogram  $I_{raw}$  is reweighted by the corresponding smoothed interferogram  $I_{smooth}$  in terms of

$$I_{\rm corr} = \frac{I_{\rm raw}}{I_{\rm smooth}}$$

The reweighting compensates the intensity fluctuations during the measurement and adjusts the modulation height in the interferogram (Fig. 1).

Keppel-Aleks et al.'s (2007) analysis is focused on the near infrared spectral region measured simultaniously with an InGaAs diode and a Si diode detector. Their full SBF correction is integrated into the software slice-ipp, and the smoothed interferogram *I*<sub>smooth</sub> is generated in three steps: (1) taking a fast Fourier Transformation (FFT) of the



(1)

raw DC interferogram, (2) applying a spectral filter to the Fourier Transform, removing all interferometric modulation, and (3) taking a second FFT of the filtered Fourier Transform. In their analysis, they show that the precision and accuracy of  $CO_2$  total column concentrations can be improved by the application of a SBF correction to spectra in the near infrared spectral region (3800–15750 cm<sup>-1</sup>).

However, in the mid infrared spectral region  $(700-3800 \text{ cm}^{-1})$  the analysis of many important traces gases, such as  $O_3$ , is fundamental and the application of SBF correction is reasonable. Between  $3800 \text{ cm}^{-1}$  and  $2000 \text{ cm}^{-1}$ , FTIR spectra are measured with an InSb detector. Below  $2000 \text{ cm}^{-1}$ , FTIR spectra are usually measured with a photoconductive MCT (mercury-cadmium-telluride) detector. Spectra, measured with such a MCT detector, are generally considered uncorrectable (Griffith and de Haseth, 1986; Rao, 1992); during the measurement process, the MCT is applied with a constant voltage, which adds an unknown offset *O* to the measured DC interferogram (Fig. 2). The unknown offset perturbs the SBF correction method (Eq. 1), since the reweighting

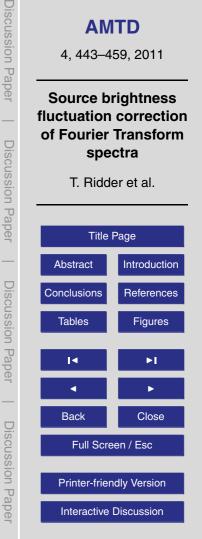
<sup>15</sup> of the raw interferogram with the smoothed interferogram now amplifies the modulation by an incorrect ratio. Thus, the unknown offset has to be removed prior to a SBF correction.

Removing the unknown offset is essential to allow for a SBF correction of MCT interferograms. However, a procedure to determine the unknown offset has not been published so far. Thus, MCT interferograms have not been corrected for SBFs in previous studies.

In Sect. 2, we present a solution to remove the unknown offset in MCT, DC interferograms allowing MCT spectra for an application of source brightness fluctuation correction. We present a method of source brightness fluctuation correction, which is independent of the measured wavelength and can, thus, be applied to spectra in the whole infrared spectral region.

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In Sect. 3, we study the impact of source brightness fluctuation correction on MCT spectra. We further investigate the influence of SBF correction on the retrieval of  $O_3$  profiles and total column concentrations. In addition, we compare our SBF correction





method to the method presented by Keppel-Aleks et al. (2007) based on the example of  $CO_2$  total column concentration measurements in Bialystok, Poland.

### 2 Method

Solar absorption FTIR spectra are obtained by a Bruker IFS 120/5M and a Bruker IFS
125HR Fourier Transform Spectrometer. The IFS 120/5M was applied during a field campaign aboard research vessel (RV) Sonne in 2009. Here, the problem of SBF had to be addressed in particular, since the campaign was planned as a North-South transit from Japan to New Zealand crossing over the tropics, where an increased appearance of clouds were expected. By default, the 120/5M measures in AC mode, but was adjusted to measure in DC mode with all detectors (InGaAs, Si, InSb, MCT) for the whole infrared spectral region. Measurements with the MCT detector were performed with low signal intensity, in order to avoid non-linearity effects. The IFS 125HR is based in Bialystok, Poland, and measures in DC mode only for the near infrared range (3800–15750 cm<sup>-1</sup>) with an InGaAs diode and a Si diode detector.

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The offset in MCT interferograms can be removed in two ways. In the first case, we measure the modulation efficiency

$$M = \frac{A}{B - O}$$

of the instrument (Fig. 2), using an InSb detector and a small optical filter. In Eq. (2), A is the AC signal intensity and B is the DC signal intensity plus the offset. The measurement is repeated with the MCT detector, using the same filter and optical settings. Thus, we can assume that the modulation efficiency is the same for both detectors

$$M_{\rm InSb} = \frac{A_{\rm InSb}}{B_{\rm InSb}} = \frac{A_{\rm MCT}}{B_{\rm MCT} - O}$$

and the offset can then be calculated as



$$O = B_{\rm MCT} - \frac{A_{\rm MCT}}{M_{\rm InSt}}$$

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The second way to remove the offset in MCT interferograms can be applied to a pair of interferograms, which is measured directly in series, if both centerbursts differ in their intensity due to SBF (Fig. 2). The modulation efficiency in each interferogram must be 5 equal

$$M_{\rm MCT_1} = \frac{A_{\rm MCT_1}}{B_{\rm MCT_1} - O} = \frac{A_{\rm MCT_2}}{B_{\rm MCT_2} - O} = M_{\rm MCT_2}$$

and the offset is calculated as

$$O = \frac{A_{\text{MCT}_2} \cdot B_{\text{MCT}_1} - A_{\text{MCT}_1} \cdot B_{\text{MCT}_2}}{A_{\text{MCT}_2} - A_{\text{MCT}_1}}$$

This method is independent of using an additional detector in comparison to the first method.

For the SBF correction (Eq. 1) all operations are performed within OPUS (version 6.5), the standard Bruker spectroscopy software. I<sub>raw</sub> is measured with the OPUS measurement procedure,  $I_{\text{smooth}}$  is generated by a direct smoothing of  $I_{\text{raw}}$  with the OPUS running mean function (mean over *n* datapoints), and the reweighting is performed by the OPUS calculator. The procedure has the advantage that the correction is independent of the measured spectral region and it can be applied automatically and instantanously together with the measurement process.

Admittedly, the OPUS running mean function still ignores datapoints at the edges of  $I_{raw}$ , and, thus, the reweighting is inaccurate there. However, this problem is of no great consequence for the procedure; in forward-backward scans the edges of the interfero-20 gram are only of importance for the phase correction (Brault, 1987; Chase, 1982), and the phase correction can be adjusted by slightly reducing the phase resolution. In single scans the problem can be avoided by minimally reducing the resolution and phase resolution of the measurement.

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#### 3 Results

#### 3.1 MCT

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Figure 3 shows the SBF correction of a solar absorption FTIR interferogram measured with the 120/5M spectrometer using a MCT detector. Three interferograms are shown

in Fig. 3 (AC, DC, SBF-corrected), as well as two spectra created from the AC and the SBF-corrected interferogram. The AC, forward-backward interferogram reveals that the measurement is influenced by SBF, causing different heights in both interferograms (Fig. 3, AC). More obviously, the influence of SBF is visible in the DC interferogram (Fig. 3, DC). The DC interferogram indicates an intensity loss towards the end of the measurement and shows the same modulation loss as in AC mode. Furthermore, the

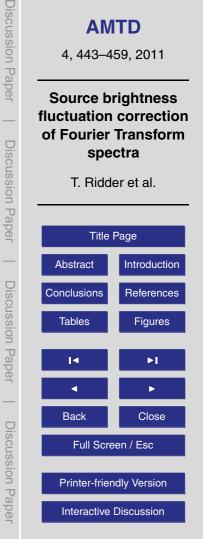
DC interferogram features the typical MCT offset.

For the SBF correction, the offset in the MCT, DC interferogram has to be removed. Following the first method (Eq. 4) the offset is calculated as O = 0.546519 using a modulation efficiency of  $M_{\text{InSb}} = 87\%$  measured with the InSb detector. Following the second method (Eq. 6), the offset is calculated as O = 0.548764, in excellent agreement to the first method (deviation of 0.41%).

Following the removal of the offset, the SBF correction with OPUS can be applied. Thereby, the phase resolution was reduced from  $4 \text{ cm}^{-1}$  to  $4.7 \text{ cm}^{-1}$  according to Sect. 2. The correction compensates the intensity fluctuations and reweights the interferometric modulation visible in the equalized heights in both interferograms (Fig. 3, SBF-corr).

The corresponding spectra of the AC interferogram and the SBF-corrected interferogram are shown in Fig. 3 (right). The SBF correction slightly improves the signal to noise ratio (SNR) of the spectrum from  $SNR_{AC} = 156$  to  $SNR_{SBF-corr} = 164$ . In addition, the procedure corrects spectral errors, visible e.g. in the spectral range between  $990 \text{ cm}^{-1}$  and  $1070 \text{ cm}^{-1}$ . Here, a variety of spectral lines are generally saturated.

990 cm<sup>-1</sup> and 1070 cm<sup>-1</sup>. Here, a variety of spectral lines are generally saturated. However, the AC spectrum shows a strong oversaturation in this range, whereby this effect is corrected in the SBF-corrected spectrum.





The influence of SBF correction on the retrieval of trace gas concentrations is demonstrated in Fig. 4, based on the example of O<sub>3</sub> retrieved from MCT, DC spectra. Three interferograms are shown, which were measured in series within one hour. The first interferogram was measured under clear sky conditions, the second and third interfer-<sup>5</sup> ogram were measured under the influence of SBF (Fig. 4, left). All three interferograms show the typical MCT offset.

The retrieval of  $O_3$  profiles and total column concentrations was accomplished in each case for the AC and the SBF-corrected spectrum, using the retrieval software SFIT2 (Rinsland et al., 1998). For the retrieval of  $O_3$ , the standard NDACC microwindow (1000–1005 cm<sup>-1</sup>) was used. Since the measurements were performed within one hour, it can be assumed that the  $O_3$  concentrations have not significantly changed during this time period.

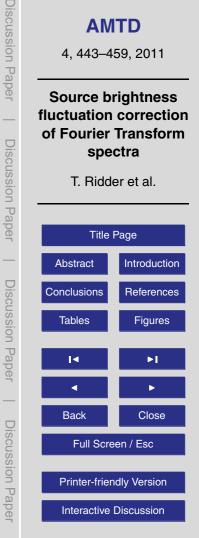
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In Fig. 4 (center) three measurements of  $O_3$  profiles are shown. The first one was performed under clear sky conditions, whereas the second and the third one were performed under the influence of SBE in the first case, the O profiles of the AC spectrum.

formed under the influence of SBF. In the first case, the O<sub>3</sub> profiles of the AC spectrum (AC) and the SBF-corrected spectrum (SBF-corr) are indentically, showing that the SBF correction has no influence on undisturbed spectra. In the second case, under the small influence of SBF, the O<sub>3</sub> profile from the AC spectrum differs from the first case. In contrast, the O<sub>3</sub> profile from the SBF-corrected spectrum equals the undis turbed profiles in case one. In the third case, the influence of SBF is more obvious. The AC, O<sub>3</sub> profile strongly differs from the original profile. However, the O<sub>3</sub> profile from the SBF-corrected spectrum equals the undis-

The impact of SBF can also be seen in the  $O_3$  total column concentrations (Fig. 4, right). In the undisturbed case the total column concentrations of the AC spectrum and

the SBF-corrected spectrum are identical (deviation of 0.13%). In the second and third case, the AC total column concentrations differ from the undisturbed case with a deviation of 1.74% and 6.73%, respectively. The SBF-corrected total column concentrations, however, equal the undisturbed concentrations with a negligible deviation of 0.41% and 0.54%, respectively.





#### InGaAs 3.2

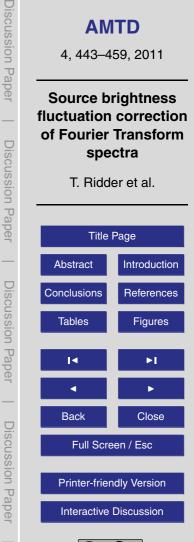
Here, we compare our SBF correction method to the method presented by Keppel-Aleks et al. (2007) for CO<sub>2</sub> total column concentration measurements in Bialystok, Poland, in April 2009. The CO<sub>2</sub> measurements were performed according to the TC-

CON standard with a 125HR spectrometer in single-scan mode. Likewise, the CO<sub>2</sub> re-5 trieval was accomplished following the TCCON approach (microwindows: 6220 cm<sup>-1</sup>. 6339 cm<sup>-1</sup>) using the retrieval software GFIT (Wunch et al., 2010; Toon et al., 1992).

The comparison contains three kinds of spectra (Fig. 5): uncorrected spectra, SBFcorrected spectra according to Keppel Aleks et al. (slice-ipp SBF-corr), and SBFcorrected spectra according to the method presented here (OPUS SBF-corr). For the 10 OPUS SBF correction the spectra were modified as discussed in Sect. 2; the resolution was reduced from 0.014 cm<sup>-1</sup> to 0.0141 cm<sup>-1</sup> and the phase resolution was diminished from  $4 \text{ cm}^{-1}$  to  $4.7 \text{ cm}^{-1}$ .

Figure 5 shows the xCO<sub>2</sub> total column-averaged dry air mole fraction (Messerschmidt et al., 2010; Washenfelder et al., 2006) for Bialystok, Poland, in April 2009 for all three cases. All three cases agree well within the intervals where the measurements are not influenced by SBF (Fig. 5, white background). In the intervals where the spectra are influenced by SBF (Fig. 5, grey background), the precision of the  $CO_2$ total column concentrations decrease for uncorrected spectra. SBF-corrected spectra, following Keppel-Aleks et al. (2007), show an improvement in the precision resulting in

20 a similar precision as for undisturbed spectra. The CO<sub>2</sub> total column concentrations from the spectra corrected with the SBF correction method presented here show the same improvement in the precision.



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#### 4 Conclusions

We showed a source brightness fluctuation correction method for solar absorption Fourier Transform infrared spectra, which is independent of the detector and wavelength and can be used for a correction of the whole infrared spectral region.

<sup>5</sup> We tested our source brightness fluctuation correction method by comparing it to the method used within the TCCON network (Keppel-Aleks et al., 2007), based on the example of  $CO_2$  total column concentration measurements. We found an improvement in the precision of  $CO_2$  total column concentrations identical to the TCCON approach.

Spectra in the spectral region below 2000 cm<sup>-1</sup> measured with a MCT detector were previously considered uncorrectable due to an unknown offset in the interferogram. We presented a solution to remove the unknown offset in MCT interferograms allowing MCT spectra for an application of source brightness fluctuation correction.

We showed a source brightness fluctuation correction applied on MCT spectra, which improves the quality of the spectra in terms of signal to noise ratio and spectral errors, resulting in an improved retrieval of  $O_3$  profiles and total column concentrations.

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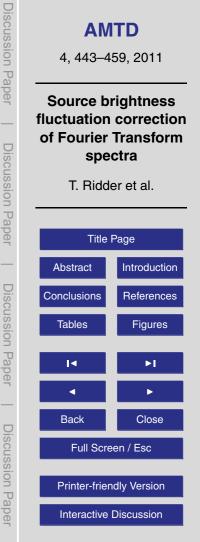
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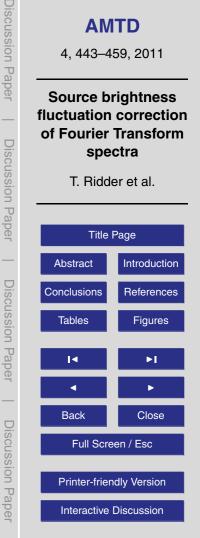
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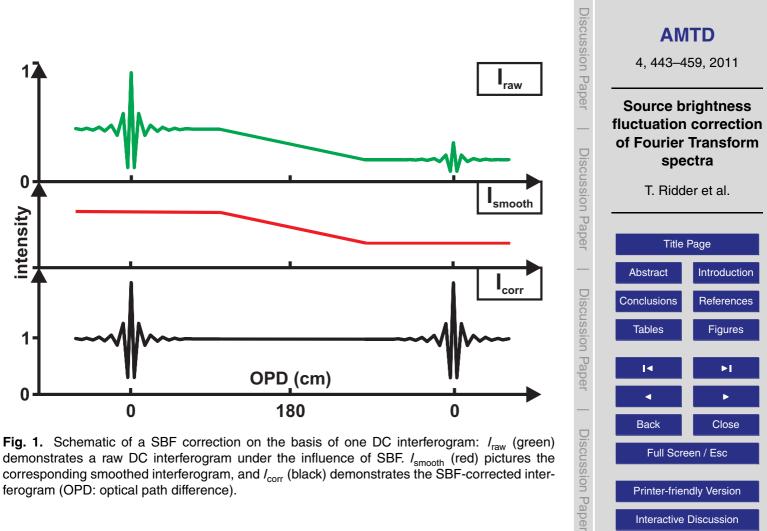
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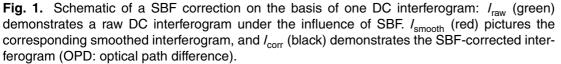




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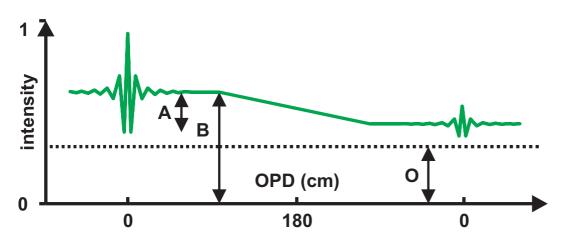




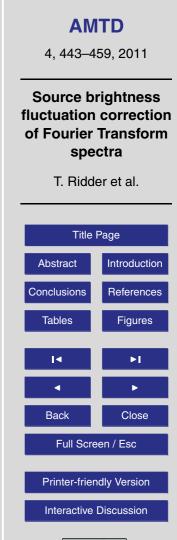
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Interactive Discussion



**Fig. 2.** Schematic of a DC interferogram measured with a MCT detector showing an unknown offset *O*. The ratio *A* (modulation height) to B-O (background intensity) demonstrates the modulation efficiency of the instrument.



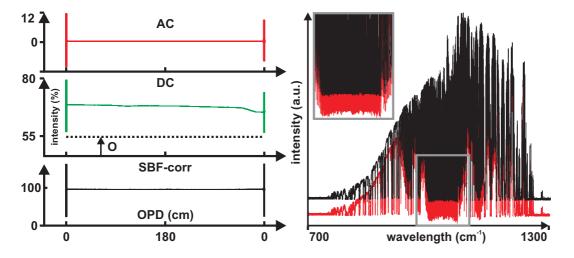
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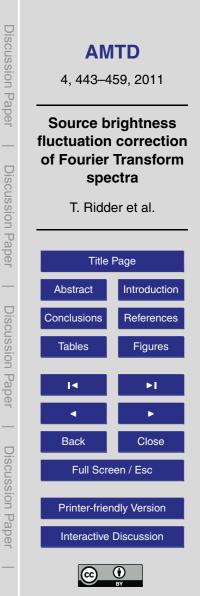
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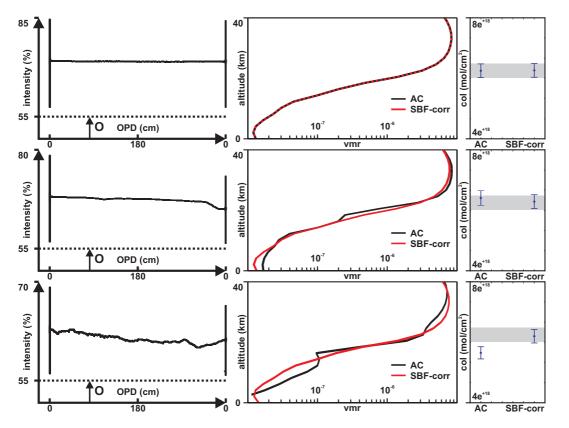
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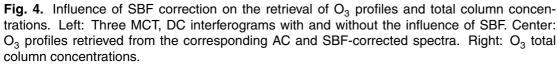


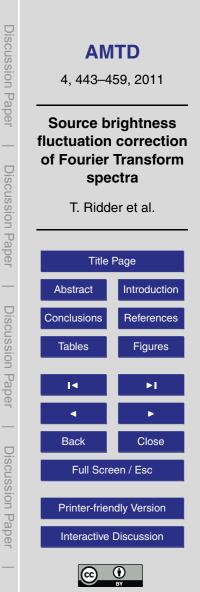


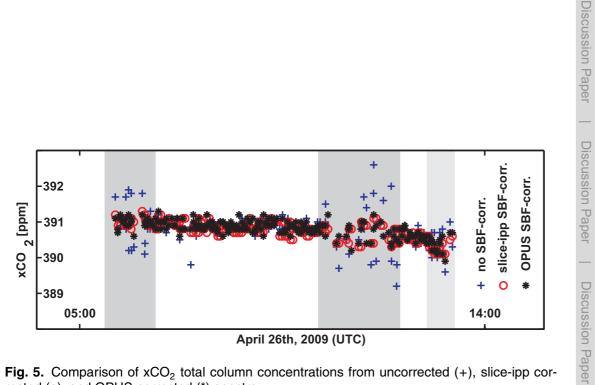
**Fig. 3.** SBF correction of a solar absorption MCT interferogram. Left: AC interferogram in comparison to DC interferogram and SBF-corrected interferogram. Right: Corresponding AC spectrum and SBF-corrected spectrum.

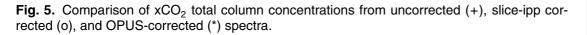














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