Use of portable FTIR spectrometers for detecting greenhouse gas emissions of the megacity Berlin – Part 1: Instrumental line shape characterisation and calibration of a quintuple of spectrometers

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Abstract

Several low resolution spectrometers were used to investigate the CO$_2$ and CH$_4$ emissions of the megacity Berlin. Before and after the campaign the instruments were tested side-by-side. An excellent level of agreement and stability was found between the different spectrometers: the drifts in $X_{\text{CO}_2}$ and $X_{\text{CH}_4}$ are within 0.005 and 0.035 %, respectively. The instrumental line shape characteristics of all spectrometers were found to be close to nominal. Cross-calibration factors for $X_{\text{CH}_4}$ and $X_{\text{CO}_2}$ were established for each spectrometer. An empirical airmass correction factor has been applied. As a last calibration step, using a co-located TCCON spectrometer as a reference, a common factor has been derived for the low-resolution campaign spectrometers, which ensures that the records are compatible to the WMO in-situ scale. Finally as a first result of the Berlin campaign we show the excellent agreement of ground pressure values obtained from total column measurements and in situ records.

1 Introduction

The continuing increase of atmospheric greenhouse gas abundances is the major driver of anthropogenic global warming. Accurate measurements of the variable atmospheric concentrations are required for the quantification of sinks and sources of these gases (Olsen and Randerson, 2004). In the last years great efforts have been undertaken to measure column-averaged dry air mole fractions of greenhouse gases with global coverage. Examples are satellite-borne instruments like SCIAMACHY (Frankenberg et al., 2006), GOSAT (Morino et al., 2011) or the recently launched OCO-2 sensor (Frankenberg et al., 2015). For the validation of OCO-2, a network of ground based high resolution Fourier-transform infrared (FTIR) spectrometers of the type 125HR from Bruker has been initiated by Caltech: the Total Carbon Column Observation Network (TCCON). Currently, about 23 TCCON globally distributed stations measure the column-averaged abundances of greenhouse gases in the atmosphere, by recording
solar absorption spectra in the near infrared (NIR) (Wunch et al., 2010). TCCON has been carefully calibrated against in situ aircraft measurements and sets the reference for remote-sensing measurements of column-averaged greenhouse gas observations. However, it is difficult to use this technical approach for the observation of sources and sinks on a regional scale, because the laboratory spectrometers applied for TCCON are not portable. Recently, KIT developed in cooperation with Bruker, Ettlingen, a portable low resolution FTIR spectrometer for the observation of greenhouse gases in the NIR and demonstrated the excellent stability of the device (Gisi et al., 2012). The spectrometer is now available from Bruker under the part name EM27/SUN. This lightweight device has low infrastructure demands so it can be operated on a campaign basis, at remote places and even on mobile platforms as ships (Klappenbach, 2015). These features not only enable the EM27/SUN to contribute to the total column measurements of the TCCON in previously underrepresented regions, in particular it can be used to gain additional information about isolated sinks and sources of greenhouse gases. Boundary layer abundances of greenhouse gases influenced by emissions from cities have been observed since long using mass spectroscopy (von der Weiden-Reinmüller et al., 2014) or cavity ring down techniques (Newman et al., 2013). The downside of this approach is the high sensitivity to local sources which overemphasizes the near vicinity and the sensitivity with respect to assumptions on vertical exchange of air masses. Here we demonstrate another approach to measure the emissions of a mega city. For this purpose, we operated five EM27/SUN spectrometer surrounding the Berlin conurbation. Over a period of three weeks, we measured the total column of CO₂, CH₄, H₂O and O₂ at the different stations. As the emission of Berlin is small compared to the atmospheric background signal, in the sub percentage order, high precision and stability of the instrument are a prerequisite. This kind of method has been applied for the quantification of individual exhausts before, however, focusing on gases which do not require a comparable level of precision and stability (Mellqvist et al., 2010). In the first part of this study, we present the comprehensive calibration procedures of the spectrometers which we applied. We performed lab-air observations
of water vapour signatures for the determination of instrumental line shape (ILS) characteristics. Moreover, we tested the participating instruments side-by-side for several days before and after the campaign, determined the level of instrumental stability and deduced calibration factors for \( X_{\text{CH}_4} \) and \( X_{\text{CO}_2} \) in order to assure that data measured by different spectrometers are compatible between each other and with TCCON measurements.

2 Instrumentation and spectrometer characteristics

2.1 EM 27 SUN spectrometer

For the acquisition of solar spectra we utilize the Bruker EM27/SUN which was developed in collaboration with the KIT. A detailed description of the spectrometer can be found in Gisi et al. (2012), in the following only a short overview including changes from the original setup is given.

The EM27/SUN features a RockSolid™ pendulum interferometer with two cube corner mirrors and a CaF\(_2\) beamsplitter. This setup achieves high stability against thermal influences and vibrations. Gimbal-mounted retroreflectors move a geometrical distance of 0.45 cm leading to an optical path difference (OPD) of 1.8 cm which corresponds to a spectral resolution of 0.5 cm\(^{-1}\). As a minor modification of the prototype spectrometer described by Gisi et al. (2012), the production device contains an off-axis mirror with a focal length of 127 mm for centering the solar beam on the detector. Together with the field stop (0.6 mm diameter) this leads to a semi Field-of-View (FOV) of 2.36 mrad. Measurements are recorded with an InGaAs detector operated at ambient temperature. Due to an electronics update it is now possible to record double-sided interferograms (IFG) of 0.5 cm\(^{-1}\) resolution. The detector is a photodiode type G12181-010K from Hamamatsu with a size of 1 mm \( \times \) 1 mm and spectral coverage from 5000 to 11 000 cm\(^{-1}\). In contrast to the detector used in the prototype that operated in the spectral region between 6000 and 9000 cm\(^{-1}\), the wider spectral coverage allows the
observation of CH$_4$. In addition, total columns of O$_2$, CO$_2$ and H$_2$O are derived from the recorded spectra. The detector signal is DC coupled and thereby supports the correction of variable atmospheric transmission (Keppel-Aleks et al., 2007).

### 2.2 Ghost to parent ratio

The EM 27 records spectra in the region from 100 to 15 798 cm$^{-1}$, so in order to satisfy the Nyquist theorem the sampling of the IFG has to be performed at every zero-crossing of the laser signal (HeNe laser, wavelength 633 nm). If the signal is not taken at exactly zero intensity, systematic sampling errors are introduced leading to artefacts in the measured spectrum, so called sampling ghosts (Messerschmidt et al., 2010; Dohe et al., 2013). Bruker recently released an effective workaround for this problem which we adopted for our measurements. A temporal linear interpolation is applied for locating the downward zero crossings. This method suppresses the ghosts below the detection limit ($< 5 \times 10^{-6}$). In addition, we tested this setup for possible line shape errors and other kinds of out-of-band artefacts, but found no detrimental effects.

### 2.3 Instrumental line shape

Precise knowledge of a spectrometer’s instrumental line shape (ILS) is of utmost importance to gain correct information from measurements as using wrong ILS values leads to systematic errors in the gas retrieval. The ILS can be divided into two parts. One part describes the modulation loss through inherent self-apodization of the spectrometer which is present also in an ideal instrument. This contribution can easily be calculated utilizing the OPD and FOV of the spectrometer. The other component of the ILS results from misalignments and optical aberrations of the spectrometer and can be characterised by a modulation efficiency amplitude and a phase error, both functions of the OPD (Hase et al., 1999). These parameters have to be deduced from lab measurements.
For the TCCON spectrometer, the standard procedure to derive the ILS are gas cell measurements. In contrast, we determine the ILS by measuring several meters of lab air and evaluating the water vapor lines in the spectral region between 7000 and 7400 cm\(^{-1}\). As light source a collimated standard 50 W halogen light bulb is used. With this approach no gas cell is necessary, which is advantageous for measurement campaigns. For the analysis of the measured data we use version 14 of the retrieval software LINEFIT (Hase et al., 1999). In LINEFIT one can choose between a simple and extended ILS model. As the ILS characteristics were close to nominal, we used the simple two-parameter ILS model. For the H\(_2\)O linelist we use the HITRAN 2009 linelist with minor adjustments, see Sect. 4.1. Needed parameters are ground pressure, ambient temperature and the distance between spectrometer and light source. Temperature and pressure were recorded using the MHB-382SD data logger with a temperature accuracy of ±0.8 °C and pressure accuracy of ±3 hPa (above 1000 hPa) or ±2 hPa (below 1000 hPa). A typical fit result is shown in Fig. 1. The SD of the residual is very low, \(1\sigma = 0.24\%\).

We measured the ILS for the different spectrometers before and after the Berlin campaign. The resulting ILS values are presented in Table 1. For the trace gas retrieval we use the mean value of the measurements before and after the campaign, the setup for these experiments was exactly the same. One can see that the values show very good agreement. The correlation between modulation efficiency amplitude and \(X_{CO_2}\) was deduced from a sensitivity study and is in agreement with Gisi et al. (2012). Instrument 2 has the biggest difference in terms of ILS modulation efficiency before and after the campaign with 0.24 %, corresponding to a change of only 0.04 % for the column-averaged dry-air mole fraction (DMF) of carbon dioxide, \(X_{CO_2}\). Note that this is not self-evident since the instruments were transported from Karlsruhe to Berlin in a Transporter, thus experiencing a lot of mechanical impacts and vibrations.
3 Measurement sites and data acquisition

In order to measure during the campaign small differences upstream and downstream of a source, a instrument to instrument consistency is of utmost importance for this setup. For this purpose, calibration measurements were carried out.

3.1 Calibration measurements at KIT Campus North

The calibration measurements were performed before the Berlin campaign on three sunny days between 6 June and 16 June and after the campaign on three consecutive days 16–18 July on top of our office building north of Karlsruhe, with an altitude of 133 m a.s.l., coordinates are 49.094° N and 8.434° E. The spectrometers were moved from the lab on the fourth floor to the roof terrace on the seventh floor thus being exposed to mechanical stress. Then they were coarsely oriented north, without effort for levelling. If further orientation was needed, we manually moved the spectrometer so that the solar beam was centered onto the entrance window. The CamTracker program was then able to track the sun. As we operated the EM27/SUN in summer, it was heated up to temperatures above 40°C. In order to protect the electronics from the heat, we built a sun cover for the EM27/SUN, which considerably reduced the temperatures inside the spectrometer. We recorded double-sided interferograms with 0.5 cm$^{-1}$ resolution. With 10 scans and a scanner velocity of 10 kHz, one measurement takes about 58 s.

For precise time recording, we used a GPS Receiver. Additionally on-site pressure and temperature profiles are available from tall tower meteorological measurements.

3.2 Berlin campaign measurements

We decided to target Berlin for several reasons. Firstly, Berlin is a megacity, so we expect to measure detectable enhancements. Secondly, the city is relatively isolated,
so that CO\textsubscript{2} emissions really can be attributed to Berlin. Thirdly, the flat topography is favorable, which supports the interpretation of the recorded data.

During the campaign period 23 June–11 July measurements were performed at five different stations around Berlin, four of them roughly located on a circle with a radius of 12 km around the city centre of Berlin. One instrument was positioned inside the Berlin motorway ring in Charlottenburg, closer to the city centre than the other instruments. A map with the different sites is shown in Fig. 2. The coordinates and altitudes of the different stations are displayed in Table 2. At the sites, temperature and pressure profiles were recorded using the MHB-382SD data logger. To obtain comparable data, we measured a long time series in Karlsruhe to determine calibration factors between the different loggers. The data was used to calculate the exact altitude of the stations. The records of ground pressure were also used in the creation of the model atmosphere. The measurement procedures (scan speed, resolution, numerical apodisation, etc.) applied during the campaign were identical to those applied for the calibration measurements.

4 Data analysis

4.1 Data processing

In a first step, the recorded interferograms are Fourier transformed using the Norton-Beer-Medium apodisation function. This apodisation is useful for reducing sibelobes around the spectral lines, an undesired feature in unapodised low resolution spectra, which would complicate the further analysis. Furthermore, a DC-correction is performed. Together with a quality filter, which discards IFGs with intensity fluctuations above 10\% and intensities below 10\% of the maximal modulation amplitude, this is implemented in a Python tool. In this work, we analyzed spectra utilizing the PROFFIT Version 9.6. This code is in wide use and has been thoroughly tested in the past, e.g. (Schneider and Hase, 2009; Schneider et al., 2010; Sepúlveda et al., 2012). Due to
the low resolution of the EM27/SUN, we fitted the atmospheric spectra by scaling of a-priori trace gas profiles. As source of the a-priori profiles, we utilized the WACCM ver. 6 climatology (http://www2.cesm.ucar.edu/working-groups/wawg).

For the retrieval we need accurate temperature and pressure profiles. In case of the Karlsruhe calibration experiments we use on-site data together with MERRA model data, which provides temperature and pressure data on a 1.25° × 1.25° grid from 1000 to 0.1 hPa 8 times a day. For the Berlin campaign we utilize local meteorological radiosonde data and the NCEP model to set up the temperature profiles. We take the NCEP data as the starting values and apply a linear ascent during the day, which is the temperature difference between the 12 a.m. and 6 p.m. sonde data, for the first height levels (until an altitude of approximately 4 km). For the height levels above 4 km we take the unaltered NCEP data, as the change during the day is negligible. For the pressure profiles we use the time series of the logger data, scale the values to 30 masl and take the smoothed mean of the different stations pressure data. We calculate the pressure of the different altitude levels from the barometric height formula using a scaling height of 8.7 km.

Every retrieval is dependent on proper spectroscopic parameters for the solar lines and atmospheric gases absorption lines. We use the HITRAN 2008 line list in its original form for CH₄, the HITRAN 2008 linelist with a line-mixing parameterisation for CO₂ adopted from a code provided by Hartmann (Lamouroux et al., 2010) and the linelist used by TCCON for O₂. For the H₂O linelist we use the HITRAN 2009 linelist with changes used by (Wunch et al., 2010) and additional ad hoc adjustments where it seemed appropriate.

### 4.2 Spectral windows

For the evaluation of the O₂ gas column we use the 7765–8005 cm⁻¹ spectral region, which is also applied in the TCCON analysis (Wunch et al., 2010). For CO₂ we subsume the spectral windows used by TCCON to one larger window ranging from 6173
to 6390 cm$^{-1}$. CH$_4$ is evaluated in the 5897–6145 cm$^{-1}$ spectral domain. For H$_2$O the 8353 to 8463 cm$^{-1}$ region is used.

An example fit for the different spectral windows is shown in Fig. 3. The residuum of the spectral fit for the water column retrieval is much bigger than for the other gases. Due to the large variability of water vapor in the atmosphere, larger linelist errors are expected. Overall the fit quality is very good with $\sigma = 0.2\%$ for CO$_2$ and CH$_4$, $\sigma = 0.1\%$ for O$_2$ and $\sigma = 0.5\%$ for H$_2$O.

5 Results calibration measurements

In this section we present results of the calibration measurements performed before and after the Berlin campaign. First we show the uncalibrated total columns followed by column-averaged DMF $X_{\text{Gas}}$ of carbon dioxide and methane, where $X_{\text{Gas}}$ is defined as

$$X_{\text{Gas}} = \frac{\text{Gas}\text{Column}}{\text{O}_2\text{Column}} \cdot 0.2095$$

To make the measurements comparable to WMO scale, in TCCON the standard procedure is to divide the calculated DMFs by a calibration factor (Wunch et al., 2010). We also apply this post processing in our work. For $X_{\text{CO}_2}$ the factor is 0.9898 whereas it is 0.9765 for $X_{\text{CH}_4}$.

5.1 Total column amounts

In Fig. 4 are depicted the column values of the measured species of the different instruments. From first glance, it is clear that the shape of all the spectrometers is nearly identical. Data gaps appearing for all instruments were caused by passing clouds. In addition Instrument 4 suffered from a hardware problem on the thirteenth of June as
well as on two days after the Berlin campaign and therefore was only partly able to perform measurements. Intraday changes of the O\textsubscript{2} column can be attributed to pressure changes for the largest part, which will be shown in Sect. 6. There are slight systematic offsets, strongest between Instrument 2 and Instrument 4 with a difference of 0.2 \%. However, note that a similar offset is also observed in the CO\textsubscript{2} and CH\textsubscript{4} gas columns, as can be seen in Fig. 4, therefore the resulting effects on the target quantities XCO\textsubscript{2} and XCH\textsubscript{4} are much smaller.

For a better comparison an intercalibration factor between the instruments is established. This is done in the following way. We take the separate measurement days and divide the data into smaller bins of 15 min duration. Inside each bin we take the mean value of all measurements from all stations and minimize the residuum for all stations. In Table 3 the calibration factors for the O\textsubscript{2} column for the calibration measurements before and after the campaign are given. Differences before and after the campaign are very small, only 0.04 \% for Instrument 2 and 4 and even less for the other instruments. This is suprisingly good, because column values are sensitive to various potential error sources, including ILS errors, timing errors, tracking errors and nonlinearities. For further analysis we use the dry air mole fraction of the gases, which are less prone to errors, because these tend to cancel out in the rationing of columns, see Eq. (1).

5.2 Column-averaged dry air mole fraction

Figure 5 shows the column-averaged DMF of CO\textsubscript{2}, which was calculated using Eq. (1). In this representation systematic errors tend to cancel out, which leads to a high degree of reproducibility in the time series difference between the instruments. Until this point, no post calibration has been performed, only the individual ILS of each instrument has been taken into account.

Table 4 shows the intercalibration factor for XCO\textsubscript{2} and XCH\textsubscript{4} before and after the campaign. The method to derive the factors is the same that was used for the O\textsubscript{2} col-
umn calibration. For $X\text{CO}_2$ we obtain a perfect agreement. The difference is below 0.005 % or 0.02 ppm for all instruments. This means we can apply a global intercalibration factor which is valid before and after the campaign. This is an important prerequisite for campaign measurements. For $X\text{CH}_4$ the agreement is slightly worse with 0.035 %, but still very good. In Fig. 6 the calibrated $X\text{CH}_4$ time series for the calibration measurements is depicted. Note that one global intercalibration factor is applied for all measurement days.

The scatter is very low for both species. Variations during the day stem from real signals, for example the $X\text{CO}_2$ peaks on the sixteenth of July were also measured by a co-located TCCON instrument (see Fig. 7).

5.3 Solar zenith angle dependency

There is a slight solar zenith angle (SZA) dependency in the $X\text{CO}_2$ and $X\text{CH}_4$ data, which is hard to see in Figs. 5 and 6 because the SZA is low during the day in summer. Also it is superimposed to the actual diurnal cycle. In order to make the data compatible with the WMO reference scale, it is nevertheless important to correct for a systematic airmass dependency originating from spectroscopic uncertainties and approximations by the radiative transfer model. For this we use a method similar to that proposed by (Wunch et al., 2010). The correction formula is

$$X_{\text{Gas}_c} = X_{\text{Gas}_{\text{unc}}} \left\{ 1 + a \left[ \left( \frac{\theta + b}{90^\circ + b} \right)^2 - \left( \frac{45^\circ + b}{90^\circ + b} \right)^2 \right] \right\}$$

where $a$, $b$ are fit parameters, $\theta$ is the SZA, $X_{\text{Gas}_c}$ and $X_{\text{Gas}_{\text{unc}}}$ are the airmass dependency corrected and uncorrected column-averaged DMF of the respective species. The choice of $45^\circ$ as the neutral angle is arbitrary and does not influence the fit results.

For the determination of the correction parameters we do not use our calibration measurement data recorded in Karlsruhe. The dataset it is not well suited for this task due to the actual intraday variability which occurred. Instead we use parameters ob-
tained from a comprehensive evaluation of EM27/SUN data from a ship cruise (Klap-
penbach, 2015). This data is not influenced by local source contributions and clearly
shows the SZA dependency in the XCO₂ and XCH₄ data. It turned out that the O₂
column-averaged mole fraction does not show a detectable SZA dependence for SZA
< 80°; the SZA dependence is essentially generated by the CO₂ and CH₄ mole frac-
tions in the numerator. The obtained parameters are \( a = 6.296 \times 10^{-3} \), \( b = 1.291 \) for
XCO₂ and \( a = 3.796 \times 10^{-3} \), \( b = 16.04 \) for XCH₄. Using this correction we receive
data comparable to in situ scale measurements, supported by the comparison with a
collocated TCCON spectrometer. Figure 7 shows the airmass dependency corrected
XCO₂ values together with TCCON DMF for 16 July. Additionally an in situ scaling was
performed to match the EM27/SUN values to the TCCON instrument. This factor of
0.99505 was determined with the method described in Sect. 5.1. The EM27/Sun val-
ues match the TCCON values remarkably well. Again one global factor was found valid
for the measurements before and after the campaign.

Figure 8 shows XCH₄ values for the same day. Similar to XCO₂ the values between
EM27/SUN spectrometers and TCCON instrument differ slightly towards evening. For
the in situ scaling factor we obtain 0.99511.

6 Berlin campaign

At the end of the first part of this study, we present the O₂ columns as recorded at all
sites during the Berlin campaign. In order to compare this dataset with in situ pressure
values derived from the MHB-382SD data loggers, we calculate the ground pressure
from the measured O₂ and H₂O total columns:

\[
P_S = \left( \frac{O_{2\text{Column}}}{0.2095 \times \bar{\mu} + H_{2O\text{Column}} \times \mu_{H_2O}} \right) \times g \times \exp \left( -\frac{\Delta h}{h_S} \right)
\]  

(2)

\( P_S \) is the surface pressure, \( \bar{\mu} \) the molecular mass of dry air, \( \mu_{H_2O} \) the molecular mass
of water, \( g \) the gravity acceleration, \( \Delta h \) the height difference between each station and
the chosen reference altitude of 30 m a.s.l. and $h_s$ the scaling height. For the sake of comparison, all pressure values derived from the spectra are transformed to this common reference altitude. We observe a systematic offset between these records and the actual ground pressure of 3.0%. This discrepancy can mainly be attributed to oxygen line intensity errors (Washenfelder et al., 2006). In Fig. 9 we scaled the pressure values obtained from the total columns to the in situ data for better comparability. The variability of the slope of the in situ measurements is nicely reproduced by the column data.

7 Conclusions

We developed a calibration procedure for mobile FTIR spectrometers which we applied to 5 spectrometers used for observing greenhouse gas emissions from Berlin during a field campaign during June and July 2014. We were successful in demonstrating the high degree of consistency. Between the instruments we established cross-calibration factors which were found valid before and after the field campaign. Drifts were below 0.005% for $X_{\text{CO}_2}$ and 0.035% for $X_{\text{CH}_4}$. In addition a method for deriving ILS parameters from open path measurements is described and was used for showing that the ILS is close to nominal for all instruments. Changes in the ILS before and after the campaign were very small, within 0.24% modulation efficiency amplitude at maximum optical path difference. Furthermore an empirical airmass correction was applied to compensate for a spurious SZA dependency of the data. As a last calibration step the in situ calibration factor derived by a comparison with a co-located TCCON instrument was applied. The same empirical calibration factor of $0.9951 \pm 0.0001$ was found valid for both $X_{\text{CO}_2}$ and $X_{\text{CH}_4}$ in order to make results comparable to WMO scale.

Finally we displayed ground pressures calculated from oxygen and water vapor columns at all sites during the Berlin campaign. The excellent station-to-station consistency and the excellent agreement with ground pressure records is a further proof of the instrumental stability.
In conclusion, we are highly confident that these portable spectrometers are very useful instruments for observing local sinks and sources of carbon dioxide and methane. In part two of this work (Hase et al., 2015), we will present the greenhouse gas observations themselves and compare these data with predictions of a simple dispersion model.

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We acknowledge the Global Modelling and Assimilation Office (GMAO) and the GES DISC for the dissemination of Merra meteorological datasets.

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References


Table 1. Compilation of ILS modulation efficiencies before and after the measurement campaign.

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<th>Instr.</th>
<th>3 Jun</th>
<th>15 Jul</th>
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</thead>
<tbody>
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<td>1</td>
<td>0.9979</td>
<td>0.9996</td>
</tr>
<tr>
<td>2</td>
<td>0.9914</td>
<td>0.9938</td>
</tr>
<tr>
<td>3</td>
<td>0.9971</td>
<td>0.9997</td>
</tr>
<tr>
<td>4</td>
<td>1.0010</td>
<td>1.0020</td>
</tr>
<tr>
<td>5</td>
<td>0.9959</td>
<td>0.9963</td>
</tr>
</tbody>
</table>
Table 2. Coordinates and altitude of the Berlin measurement stations.

<table>
<thead>
<tr>
<th>Site</th>
<th>Long. (° E)</th>
<th>Lat. (° N)</th>
<th>Altit. (m a.s.l.)</th>
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<tr>
<td>Mahlsdorf</td>
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<td>52.505</td>
<td>47.7</td>
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<td>Lichtenrade</td>
<td>13.392</td>
<td>52.391</td>
<td>44.8</td>
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**Table 3.** Calibration factors for O$_2$ column before and after the campaign for the different instruments. Instrument 1 has been scaled to one, which is an arbitrary choice.

<table>
<thead>
<tr>
<th>Instr.</th>
<th>O$_2$ col. before</th>
<th>O$_2$ col. after</th>
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<td>1</td>
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<td>1.00000</td>
</tr>
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<td>2</td>
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<td>4</td>
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<tr>
<td>5</td>
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<td>1.00110</td>
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</tbody>
</table>
**Table 4.** Calibration factor for $X_{CO_2}$ and $X_{CH_4}$ for the different instruments before and after the campaign.

<table>
<thead>
<tr>
<th>Instr.</th>
<th>$X_{CO_2}$ bef.</th>
<th>$X_{CO_2}$ aft.</th>
<th>$X_{CH_4}$ bef.</th>
<th>$X_{CH_4}$ aft.</th>
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</thead>
<tbody>
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<td>1.00000</td>
<td>1.00000</td>
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</tbody>
</table>
**Figure 1.** Transmission spectrum of 4 m lab air (black curve) in the 7000 to 7400 cm\(^{-1}\) region. Overlying is the LINEFIT calculation (red curve), the residuum multiplied by a factor of ten is shown in blue. For clarity reasons, an offset of −0.1 was added to the residuum.
Figure 2. Map with the Berlin measurement stations.
Figure 3. Spectral windows used during the retrieval for the different species. The fit is in accordance with the measurement, the residuum, which has been multiplied with a factor of 10 for H\textsubscript{2}O and 20 for the other species, is small.
Figure 4. Total columns of O$_2$, CO$_2$ and CH$_4$ for the different spectrometers on four days of the calibration measurements in Karlsruhe. The first two days are before the Berlin campaign, the other days after the campaign. One data point consists of 10 interferograms, the measurement time being 58 s each.
Figure 5. Uncalibrated $X_{CO_2}$ values for all instruments. Experiments and measurement days are the same as for $O_2$. 
Figure 6. Calibrated XCH₄ values for all instruments. Experiments and measurement days are the same as for O₂.
**Figure 7.** Calibrated and SZA corrected XCO$_2$ values for all EM27/SUN spectrometers on 16 July. Golden dots show XCO$_2$ data from a co-located TCCON instrument.
Figure 8. Calibrated and SZA corrected $XCH_4$ values for all EM27/SUN spectrometers on 16 July. Golden dots show $XCH_4$ data from a co-located TCCON instrument.
Figure 9. In situ pressure data together with pressure data calculated from total column amounts of $\text{O}_2$ and $\text{H}_2\text{O}$. The column data is scaled with an in situ factor for better comparability.