



Improved instrumental line shape monitoring for the ground-based, high-resolution FTIR spectrometers of the Network for the Detection of Atmospheric Composition Change

F. Hase

Karlsruhe Institute of Technology (KIT), Institute for Meteorology and Climate Research (IMK-ASF), Germany

Correspondence to: F. Hase (frank.hase@kit.edu)

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Abstract. We propose an improved monitoring scheme for the instrumental line shape (ILS) of high-resolution, ground-based FTIR (Fourier Transform InfraRed) spectrometers used for chemical monitoring of the atmosphere by the Network for Detection of Atmospheric Composition Change (NDACC). Good ILS knowledge is required for the analysis of the recorded mid-infrared spectra. The new method applies a sequence of measurements using different gas cells instead of a single calibration cell. Three cells are used: cell C1 is a refillable cell offering 200 mm path length and equipped with a pressure gauge (filled with 100 Pa N_2O), cells C2 and C3 are sealed cells offering 75 mm path length. C2 is filled with 5 Pa of pure N_2O . Cell C3 is filled with 16 Pa N_2O in 200 hPa technical air, so provides pressure-broadened N_2O lines. We demonstrate that an ILS retrieval using C1 improves significantly the sensitivity of the ILS retrieval over the current calibration cells used in the network, because this cell provides narrow fully saturated N_2O lines. The N_2O columns derived from C2 and C3 allow the performance of a highly valuable closure experiment: adopting the ILS retrieved from C1, the N_2O columns of C2 and C3 are derived. Because N_2O is an inert gas, both columns should be constant on long timescales. Apparent changes in the columns would immediately attract attention and indicate either inconsistent ILS results or instrumental problems of other origin. Two different cells are applied for the closure experiment, because the NDACC spectrometers observe both stratospheric and tropospheric gases: C2 mimics signatures of stratospheric gases, whereas C3 mimics signatures of tropospheric gases.

1 Introduction

Today, various techniques contribute to the effort of monitoring the atmosphere's chemical composition. Among these techniques, high-resolution ground-based FTIR (Fourier Transform InfraRed) spectrometers significantly contribute with accurate long-term measurements of total columns and mixing ratio profiles as function of altitude for various chemical species. The direct result of the solar FTIR measurement is a transmission spectrum of the terrestrial atmosphere. The desired target quantities are determined by adjusting the input parameters of a calculated synthetic spectrum in order to reproduce appropriate spectral sections of the observed spectrum (Hase et al., 2004). The FTIR technique allows the recording of solar spectra with high signal-to-noise ratio and achieves both wide spectral coverage and high spectral resolution. The wide spectral coverage permits the monitoring of many different species, and the high spectral resolution allows the shapes of individual spectral lines to be resolved. Thereby, due the altitude-dependent pressure broadening, not only the total column of a trace gas can be determined; in addition a retrieval of mixing ratio profiles (with moderate vertical resolution) can be achieved for many trace gases. Accurate knowledge of the instrumental line shape (ILS) is required to interpret the observed line shapes in a correct manner, and the ILS can be a significant error source in the overall error budget (Schneider and Hase, 2008).

In order to guarantee consistent results between different FTIR stations, these are affiliated with networks which supervise the compliance of instrumental setups, define the recommended practice for the analysis of the spectra and collect the results into databases which form the interfaces to the data users. The NDACC (Network for Detection of Atmospheric Composition Change; Kurylo, 1991; <http://www.ndacc.org/>)

started operation in 1991. The NDACC FTIR spectrometers provide time series for O_3 , HNO_3 , HCl , HF , CO , N_2O , CH_4 , HCN , C_2H_6 and ClONO_2 derived from mid-infrared (MIR) spectra covering the 750 to 4200 cm^{-1} spectral range. The Total Carbon Column Observing Network (TCCON, Wunch et al., 2011) is a network of high-resolution near-infrared (NIR) spectrometers which started operation in 2004. TCCON provides data for CO_2 , CH_4 , N_2O , HF , CO , H_2O , and HDO derived from NIR spectra.

It has become a part of FTIR network practice to regularly use a low-pressure calibration gas cell (either in the solar beam or using a lab source) to derive the ILS from the shape of the narrow spectral lines supplied by the cell. This retrieved ILS is used to diagnose a misalignment of the spectrometer and to improve the alignment when indicated. Moreover, the actual ILS derived from the cell measurement can be introduced into the analysis process of the atmospheric spectra, e.g. Schneider et al. (2008).

New upcoming scientific challenges, as for example the inversion of measurements to derive sources and sinks for long-lived greenhouse gases, and the associated advance of satellite data (e.g. Butz et al., 2011) and model evaluations (e.g. Chevallier et al., 2011) exert a steady pressure on the precision achieved by ground-based FTIR measurements. Currently the NDACC investigators strive for sub-percent precision of their data products (e.g. Schneider and Hase, 2008; Sussmann et al., 2011). The target precision and accuracy for TCCON's XCO_2 data product is 0.1% (Deutscher et al., 2010; Messerschmidt et al., 2011).

We believe that in order to keep up with the increasing precision and accuracy requirements, the current ILS monitoring scheme used within the NDACC needs to be further refined. In the following sections we describe the current practice of ILS monitoring in detail. Next we introduce the proposed ILS monitoring scheme and provide indication that the new setup allows for a significantly improved ILS determination and ensures network consistency.

2 Current state of the art for ILS monitoring

Regular ILS monitoring using gas cells was introduced at some NDACC FTIR stations in 1996. To achieve sufficiently narrow spectral features in the MIR, heavy molecules at pressures below 250 Pa are applied. Hase et al. (1999) used refillable N_2O cells of 20 cm path length equipped with NaCl windows at a filling pressure of 10 Pa . If appropriate window material (e.g. NaCl , KBr , KCl) is applied, this type of cell can also be used to monitor the HgCdTe detector branch, which covers the long-wavelength spectral region (750 – 1300 cm^{-1}), in addition to the short-wavelength InSb detector branch of the FTIR. Permanently sealed HBr cells with sapphire windows were provided by the National Center for Atmospheric Research, Boulder, Colorado, to the FTIR stations of the NDACC from 2000 onwards (Goldman et al.,

2003, and references therein). With a path length of 2 cm , these cells are extremely compact, they are filled with 200 to 250 Pa of HBr and have become the network standard. The sealed HBr cells are intended to be used not only for the ILS determination, but also for closure experiments: the analysis of the cell spectrum provides both the ILS and the gas column inside the cell, which should be constant for a sealed cell. Unfortunately, this highly desirable functionality has not fully been achieved, as the reactive HBr tends to decay in the cell, likely due to a chemical reaction with residual contaminations of the cell walls. In consequence, HBr does not provide a stable reference column on longer timescales. TCCON applies sealed cells of 10 cm path length filled with 500 Pa of HCl (Washenfelder, 2006), which also tends to decay on longer timescales.

For the analysis of the cell spectra, the LINEFIT code developed by Hase (Hase et al., 1999) is used by NDACC and TCCON. LINEFIT determines the ILS by performing a constrained fit of parameters which describe the interferometer's modulation efficiency in the interferogram domain. This complex modulation efficiency is defined as the ratio of the normalized modulation of the actual Fourier transform spectrometer (FTS) and the normalized modulation of a nominal FTS as function of optical path difference. The term “normalized” indicates that the modulation amplitude at zero path difference (ZPD) is normalized to unity to ensure the area-normalization of the ILS in the spectral domain. This definition is appropriate, as we are not aiming at a measure of the absolute efficiency of the spectrometer, but at characterising the ILS, which is determined by the variations of modulation efficiency as function of optical path difference (OPD). For the nominal FTS, we assume that the spectrometer only suffers from the self-apodisation effect due to the finite field-of-view of the interferometer and the limited optical path difference (e.g. Davis et al., 2001, Chapter 5.1 and 5.2), but otherwise works perfectly. To achieve an efficient formulation of constraints, it is appropriate to represent the complex modulation efficiency by its amplitude and phase. LINEFIT retrieves modulation efficiency amplitude and phase at 20 equidistant OPD positions up to the maximum value of OPD applied in the measurement. This provides 39 adjustable variables to fit any reasonable deviation from the nominal ILS (39 instead of 40 parameters, because the modulation amplitude at zero OPD is kept fixed to unity). According to the aforesaid, LINEFIT will determine unity amplitude and zero phase error along the whole interferogram, if the spectrometer under investigation behaves ideally. In addition, auxiliary parameters as e.g. scaling and slope parameters to describe the background continuum, the gas column, spectral scale and gas temperature can be fitted. LINEFIT results have successfully been compared with other approaches, e.g. ILS deconvolution (Bernardo and Griffith, 2005).

3 Limitations of the current ILS monitoring scheme

All TCCON sites and nearly all NDACC FTIR sites apply the same type of commercial spectrometers manufactured by Bruker (<http://www.brukeroptics.com>), the spectrometer type 125HR (several NDACC sites use its precursor 120HR or the 120M, which basically all share a common optical design). The heart of the spectrometer is a classical Michelson-type interferometer with a linearly moving scanner. Instead of plane mirrors, Bruker applies cube-corner reflectors in both arms of the interferometer. As a result, the interferometer is tilt- but not shear-compensated, so a lateral displacement between the cube-corners will affect the ILS (Kaupinen and Saarinen, 1992a). Especially, a lateral displacement between moving and fixed cube corner at zero optical path difference (ZPD) will reduce the modulation amplitude in the inner part of the interferogram, causing an apparent increase of modulation amplitude as function of OPD. In the general case, the lateral displacement will be a function of OPD, because the scanner arm might be tilted with respect to the optical axis defined by the interferometer's field-of-view (FOV) and might not be perfectly straight. Moreover, the FOV might be blurred due to optical misalignment, introducing a modification of the expected self-apodisation (Kaupinen and Saarinen, 1992b). Finally, finite divergence and misalignment of the HeNe reference laser might slightly distort the sampling positions, thereby impacting the phase error. Overall, no straightforward correspondence between ILS and physical misalignment exists, instead the resulting deviations from the nominal modulation efficiency result from a superposition of various small instrumental imperfections. As a consequence, a direct fit of physical instrumental misalignment parameters is difficult, and use of a prescribed ILS parameterisation using only a few parameters might be reasonably successful for one misalignment scenario but fail for another. Consequently, the most reliable approach remains the retrieval of an extended set of ILS parameters as described above.

The statements of the previous section require the following clarification to avoid a misunderstanding: the FTIR spectrometer is a highly precise and stable measurement device and the quality of spectra in practice is astounding. As outlined in the introduction, it is the ambitious sub-percent precision level the FTIR community is aiming at which draws our attention to tiny imperfections of the spectrometer. If the spectrometer's environment is well-controlled, then the main driver for a changing ILS usually is the wear of the of the moving cubecorner's friction bearing, which induces a vertical offset of the cubecorner. The modulation efficiency amplitude of a 125HR spectrometer used for regular solar measurements will typically change by a few percent on timescales of a year (at a maximal optical path difference of 180 cm). Unless the spectrometer is realigned, it can be assumed that the ILS is a slowly varying instrumental feature, so a 1–3 month cycle for the ILS characterisation

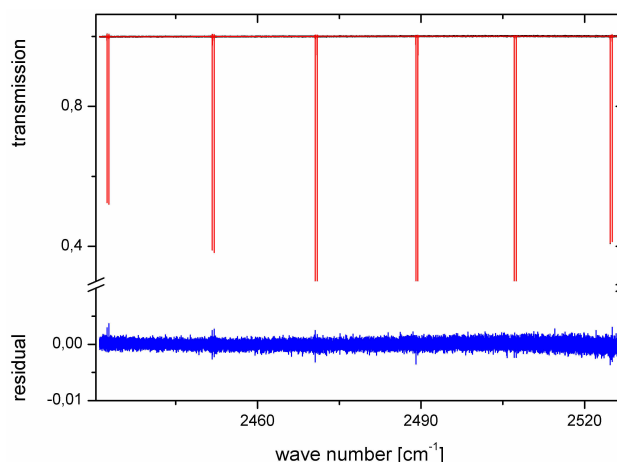


Fig. 1. Transmission spectrum recorded with the HBr NDACC cell #44 (black) and superimposed on it the LINEFIT calculation (red). The fit residual is shown in blue. The measurement has been recorded with the TCCON FTIR spectrometer in Karlsruhe on 10 November 2011.

should suffice. Figure 1 shows the spectral scene provided by the HBr cell described above and a LINEFIT calculation superimposed on it. Figure 2 shows the modulation efficiency derived from this spectrum, and Fig. 3 the averaging kernels for the retrieved ILS parameters. The sensitivity of the retrieval slightly decreases towards maximum OPD, which is not critical, because the atmospheric spectra will hardly contain spectral structures as narrow as those provided by a low-pressure gas cell. However, the sensitivity also drops significantly towards zero path difference (ZPD), which is a substantial problem. The difficulty in reconstructing the modulation efficiency in the vicinity of ZPD is due to the fact that the associated spectral features are broadband, so they mix with inevitable tiny variations of the background continuum surrounding the line. For illustration, Fig. 4 shows the resulting changes in a simulated spectrum when the modulation amplitude decreases in the inner part of the interferogram towards ZPD. Due to the over-modulation assumed in this example, the ILS peak height increases, and the conservation of ILS area is provided by associated subtle changes of the ILS sidelobes. Due to the increased ILS peak height, the absorption lines appear somewhat deeper. In case of an unsaturated spectral line, this additional depression can be misinterpreted by assuming a higher gas column, so the quantification of the modulation efficiency change between the inner and outer part of the interferogram ultimately depends on a marginal change of the observed line wings (in our example of 10 % modulation efficiency rise generates a mere 1 % signal in transmission in the near-wing region which becomes undetectable further away from the line). The HCl cell mentioned above, which is used within the TCCON network, provides unsaturated spectral lines, too, and therefore suffers from the same limitations.

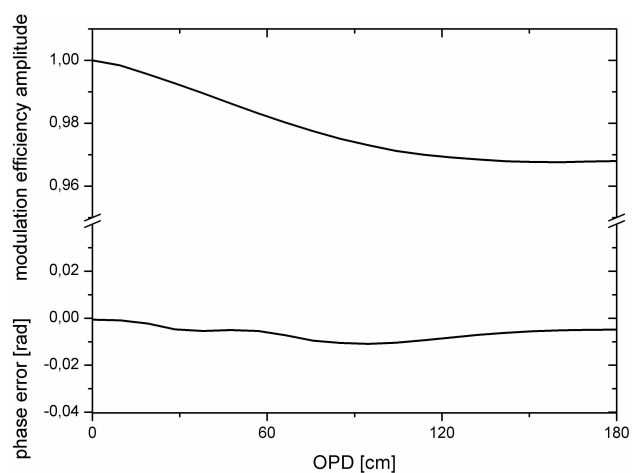


Fig. 2. The normalized modulation efficiency and phase error derived from the HBr cell spectrum shown in Fig. 1.

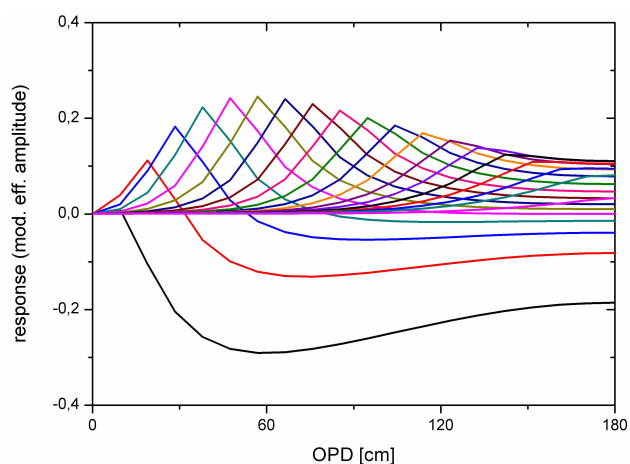


Fig. 3. The averaging kernels of the ILS retrieval from the HBr cell (the modulation amplitude kernels are shown).

4 Description of the new ILS retrieval scheme

We now demonstrate that in order to reduce the ambiguity demonstrated in the last section, the spectral scene used for the ILS retrieval should offer saturated absorption lines. Both the HBr cells used by NDACC and the HCl cells used by TC-CON achieve center absorption depths for the strongest spectral lines of about 30 % transmission. An imaginary modification of the gas column will change this residual transmission. In contrast, the depth of a saturated line generated at low pressure cannot be modified to the same extent by adjusting the assumed gas column (it is a near boxcar-shaped transmission profile which reaches the zero baseline), so the retrieval is less prone to the described crosstalk between retrieved gas column and apparent line depth changes connected to modulation efficiency. A cell with 200 mm path length filled with 100 Pa N_2O (cell C1) generates a useful

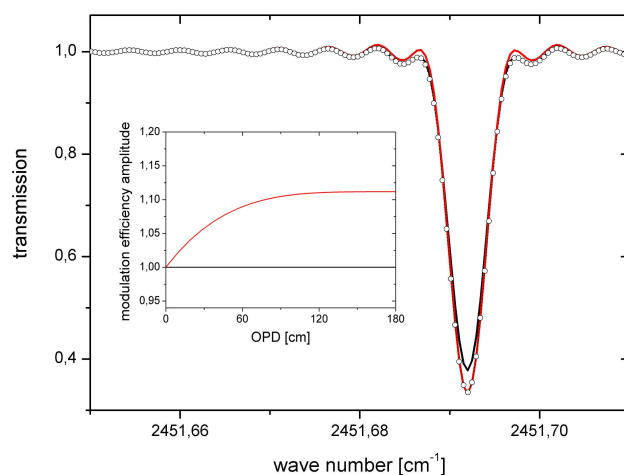


Fig. 4. The changes in an HBr absorption line which result from a 10 % over-modulation. This example strongly exaggerates the effect of a poor ZPD alignment. The black curve is the simulation with a nominal ILS, the red curve refers to the over-modulation, the open circles show a spectrum calculated with nominal ILS, but increasing the gas column by 12 %. The inserted panel shows the assumed modulation amplitude.

spectral scene which contains both saturated and unsaturated lines. Figure 5 shows the transmission spectrum provided by such a cell and a LINEFIT calculation superimposed on it. Figure 6 shows the modulation efficiency derived from this spectrum and Fig. 7 the averaging kernels for the retrieved ILS parameters, which demonstrate the improved sensitivity of the modulation efficiency retrieval in the inner part of the interferogram (to be compared with the HBr kernels shown in Fig. 3). At a first glance, it is surprising that the N_2O kernels in the outer part of the interferogram are not significantly poorer than the HBr kernels, given the considerably higher mass of HBr molecules (HBr/ N_2O mass ratio 1.8). However, the N_2O band used resides at somewhat lower wave numbers than does the HBr band; for this reason the Doppler-broadened HBr line width is only 20 % smaller than for N_2O . Moreover, working at a pressure of 240 Pa (HBr cell) and 100 Pa (N_2O cell), respectively, the pressure broadening is not fully negligible. Finally, the N_2O spectrum contains more and stronger spectral lines per wave number increment than does the HBr spectrum (compare Fig. 1 and Fig. 5), therefore more input signal is provided for the ILS retrieval, which results in a similar sensitivity of the N_2O retrieval in the outer part of the interferogram for a given signal-to-noise ratio of the spectrum.

Cell C1 is in principle sufficient for the ILS determination, but by adding two further cells – cell C2 (sealed, filled with 5 Pa N_2O) and cell C3 (sealed, filled with 16 Pa N_2O in 200 hPa technical air) – we can perform a highly valuable closure experiment. Both cells provide unsaturated N_2O lines which allow an accurate column determination. Unlike

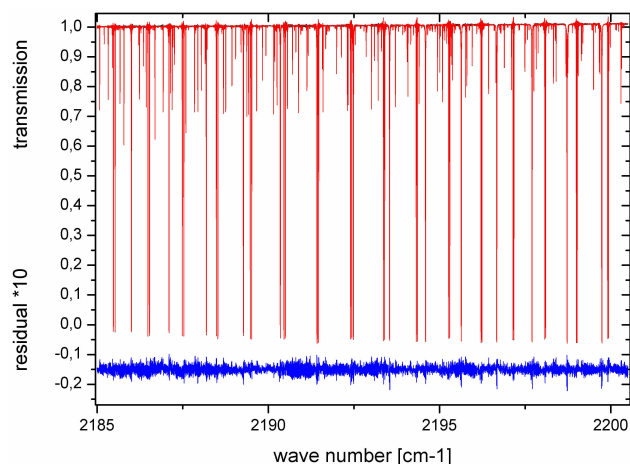


Fig. 5. A cell with 200 mm effective path length filled with 100 Pa N_2O (cell C1) generates a useful spectral scene which contains both saturated and unsaturated lines. The figure shows the transmission spectrum provided by such a cell (black curve) and a LINEFIT calculation superimposed on it (red curve). The residual is shifted and magnified by a factor of 10 (blue curve). The spectrum was recorded on 26 July 2011.

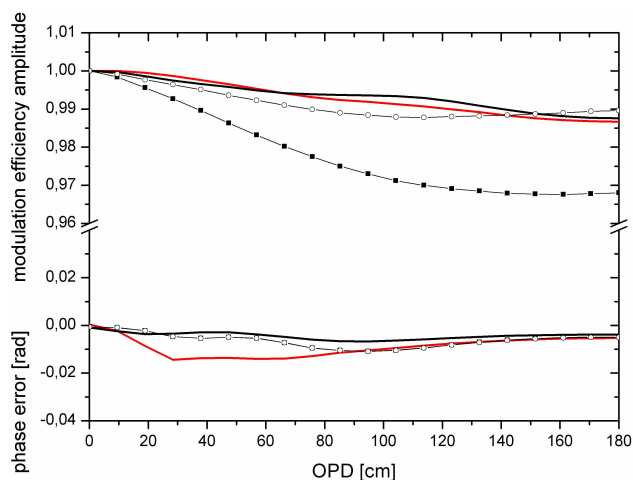


Fig. 6. The normalized retrieved modulation efficiency and phase error for July (black) and October (red) as derived from the cell C1; only a moderate change of the ILS is indicated over the three-month period. Curves with symbols: modulation efficiency derived from the HBr cell in early November (black squares: assuming nominal pressure of 240 Pa, open circles: assuming 280 Pa total pressure).

HBr or HCl, N_2O is an inert gas, therefore both columns should be constant on long timescales. We assume that the ILS is known from the analysis of the C1 measurement, so the ILS parameters are kept fixed in the analysis of the C2 and C3 spectra, only the gas column and auxiliary parameters are fitted. C2 provides narrow lines to emulate the line shape of a stratospheric species (e.g. O_3), whereas C3

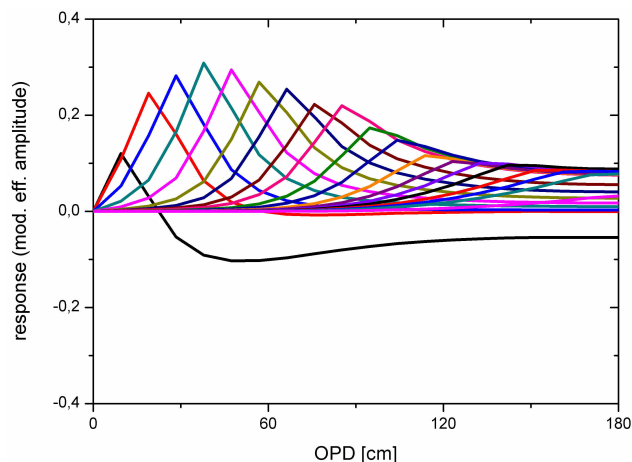


Fig. 7. The averaging kernels of the ILS retrieval for the N_2O cell spectrum shown in Fig. 5 (the modulation amplitude kernels are shown). A comparison with Fig. 3 reveals the improved sensitivity of the retrieval for small OPD.

provides pressure-broadened lines to emulate the line shape of a tropospheric species (e.g. CH_4 , N_2O). We require that the retrieved gas columns of the sealed cells C2 and C3 remain constant. Any changes of the C2 or C3 columns would immediately attract attention and indicate either inconsistent ILS results or instrumental problems of other origin.

The reader might wonder why we introduce C2 in addition to C1 for the closure experiment. The reason is that we prefer a refillable cell with an attached manometer for the ILS determination itself, so that any pressure increase due to leaking or degassing is noted immediately. If a single sealed low-pressure cell would be used for both purposes (ILS retrieval and column check), then a defect of manufacturing of this cell (rising pressure due to leakage or degassing) would interfere with the ILS determination. By keeping these two functionalities separated, we can readily uncover even a slight pressure rise in C2.

5 First results using the proposed method

We applied the new setup on a ground-based high-resolution FTIR spectrometer located north of Karlsruhe (49.100° N, 8.438° E), which is an accredited TCCON spectrometer. It successfully participated in the IMECC calibration campaign for the European TCCON sites (Messerschmidt et al., 2011). The spectrometer is equipped both with a room-temperature InGaAs detector for NIR observations and with a liquid-nitrogen cooled InSb detector and the NDACC optical filter set for MIR observations. Regular NIR and MIR solar measurements were started in September 2009. We performed cell measurements of the proposed kind on 26 July and 22 October 2011.

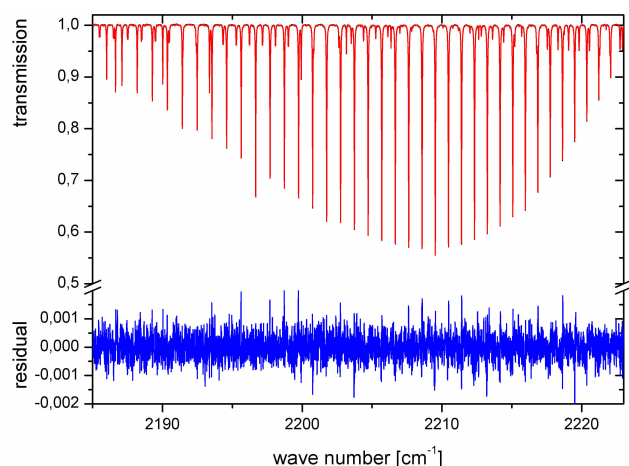


Fig. 8. The spectrum taken with cell C3 in July (black), superimposed the synthetic spectrum fitted by LINEFIT is shown (red), blue: residual.

Figure 6 compares the retrieved modulation efficiency for both dates as derived from C1, only a moderate change of the ILS has occurred over the three months period. The modulation efficiency derived from the HBr cell on 10 November is also included. All results diagnose a well-controlled phase error below 20 mrad; the October measurement indicates a slightly larger ILS asymmetry than the measurements in July and November. The HBr measurement suggests a stronger modulation loss by about 2 % towards larger OPD than do the N₂O measurements. We do not expect such a change of the ILS within two weeks. However, the HBr result depends on the assumed cell pressure in the sealed cell, assuming a total pressure of 280 Pa instead of 240 Pa in the cell would bring the results in much closer agreement. For the fits of the C2 and C3 spectra discussed in the following, we adhere to the ILS parameters of the particular date as derived from the C1 spectra. Figure 8 shows the spectrum taken with C3 on July and the synthetic spectrum fitted by LINEFIT (the C3 spectra recorded in July and October are hardly distinguishable, therefore we do not show the results for October). The derived columns are in excellent agreement: 3.036×10^{20} molec m⁻² for the July measurement and 3.027×10^{20} molec m⁻² for the October measurement. In case of C2, we detect a clear indication of a problem with this cell, the observed line shapes differ significantly between July and October, see Fig. 9. However, if we allow for a pressure rise, excellent fits to the measured spectra are achieved. Figure 10 shows the spectrum taken with C2 in July and the synthetic spectrum fitted by LINEFIT, Fig. 11 provides the equivalent information for the spectrum taken with C2 in October. The total cell pressure in C2 is in accordance with the assumption of purely Doppler-broadened line shapes in July but increased to 220 Pa in October. The retrieved columns are in excellent agreement: 8.764×10^{19} molec m⁻² for the

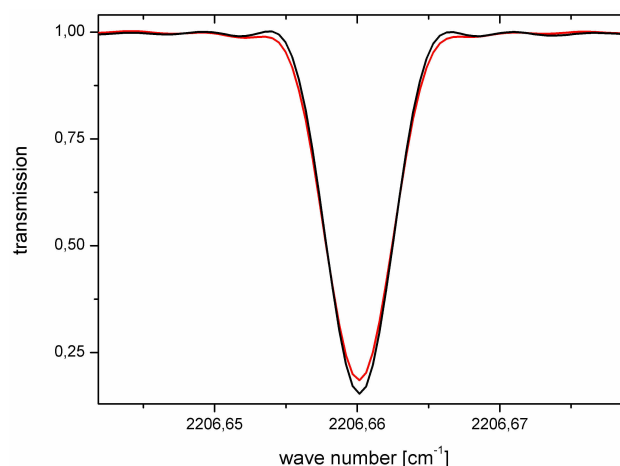


Fig. 9. Small spectral section out of the spectra recorded with the sealed cell C2 in July (black) and October (red). Although the modulation efficiency did not change significantly, the lines are significantly broader in October, indicating a pressure rise in C2.

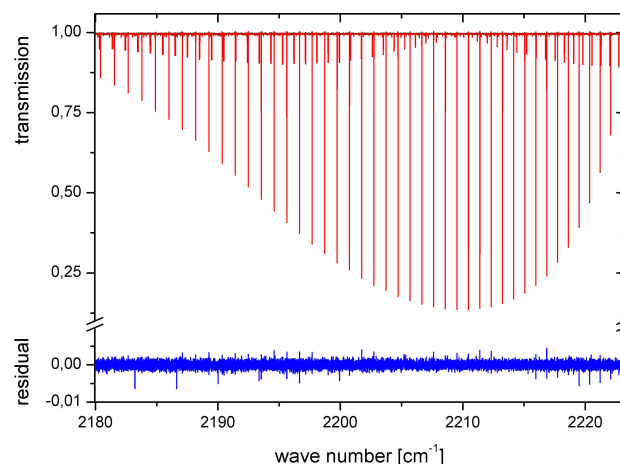


Fig. 10. The spectrum taken with C2 on 26 July 2011 (black), superimposed the synthetic spectrum fitted by LINEFIT (red) is shown, blue: residual.

July measurement and 8.721×10^{19} molec m⁻² for the October measurement.

Although several cells are involved, practical experience reveals that the effort to perform the proposed method is quite manageable. To generate the C1, C2, and C3 spectra, we co-added 20 scans and applied 180 cm maximum OPD for C1 and C2, and 45 cm OPD for the measurement of the pressure-broadened C3 spectrum. The whole set of measurements can be recorded within a few hours. Some details of our new method depend on the instrumental setup, which is not fully identical at all NDACC FTIR sites: some spectrometers offer an evacuable probe chamber inside the spectrometer for the cell. In our case, the cell was located outside the spectrometer in lab air. For this reason, we applied an

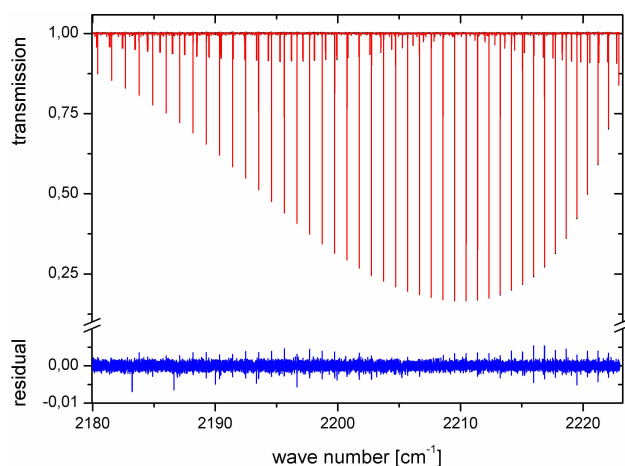


Fig. 11. The spectrum taken with cell C2 on 22 October 2011 (black), superimposed the synthetic spectrum fitted by LINEFIT is shown (red), blue: residual.

additional evacuated cell C4 identical to C3 and C2, and ratioed the C2 and C3 spectra over the spectrum taken with C4 to remove residual weak N_2O signatures from the lab air (for C1, we corrected the lab air transmission according to the longer cell). However, we feel that the use of C4 is not mandatory, the lab air transmission can be characterised by a measurement without a cell, the column of lab-air N_2O can be determined from such a measurement with LINEFIT and taken into account afterwards in the LINEFIT analysis of the cell measurements (taking into account the appropriate lab-air path).

6 Challenges and considerations for ILS monitoring within TCCON

As mentioned in Sect. 3, the current ILS monitoring scheme for TCCON suffers from the same limitations as the current NDACC scheme. Unfortunately, molecular absorption strengths are considerably weaker in the NIR than in the MIR spectral region. Due to the increasing width of the Doppler core as function of wave number it is possible to use higher cell pressure, but even if this advantage is exploited a cell body of impracticable length is required for the generation of saturated lines. N_2O , which is well suited for MIR purpose does not provide absorption bands of sufficient strength in the NIR spectral domain. Multi-path cells are common for lab spectroscopy applications, but the optical coupling of such a cell is complicate and prone to misalignment effects which might affect the ILS result. The use of NIR lasers or thermal emission line sources might turn out as advantageous, but considerable research is required to identify a manageable and reliable solution.

In contrast to NDACC, TCCON is restricted to the high-precision-and-accuracy measurement of greenhouse gases. These gases are prevalent in the troposphere and their atmospheric absorption signatures are significantly pressure-broadened. Therefore, exchange of a single high-pressure cell (equivalent to the cell C3 introduced above) between TCCON sites would be sufficient to verify the consistency of retrieved columns. One might argue that the determination of the atmospheric O_2 column from the $1.26\text{ }\mu\text{m}$ band, which is part of the TCCON analysis procedure, is sufficient for this purpose, but measuring the same gas column in a test cell across the network would nevertheless add a very convincing demonstration for the proper characterisation of all spectrometers. Whereas the O_2 column is exploited to compensate for other problems of the whole experimental setup, e.g. incorrect assumptions on observed air mass due to pointing errors, an incorrect cell column deduced from a lamp measurement would indicate a specific problem with the spectrometer itself. A cell of 50 mm effective path length filled with 200 hPa of pure CH_4 would provide lines of useful absorption strength around 6000 cm^{-1} , near to the relevant CO_2 bands.

7 Summary and outlook

The new ILS monitoring recipe provides improved sensitivity and reliability for the retrieval of ILS parameters. The ILS parameters are derived from the transmission spectrum of a refillable, pressure-monitored cell with 20 cm path length filled with 100 Pa N_2O . In the second step of the procedure, a closure experiment ensuring the integrity of retrieved ILS is performed: for this purpose the N_2O columns of two additional sealed calibration cells of 75 mm path length are monitored. In the analysis of these cells, the ILS derived from the C1 measurement is applied. One cell is filled with 5 Pa N_2O to emulate narrow spectral lines of a stratospheric absorber, the other cell is filled with 16 Pa N_2O in 200 hPa technical air to emulate pressure-broadened lines of a tropospheric absorber. First measurements with the Karlsruhe TCCON FTIR spectrometer indicate excellent repeatability of ILS and column results and show that the use of several cells is practicable. The procedure revealed a technical problem with the sealed low pressure test cell C2.

We do not think that the current HBr cell recipe is obsolete, as it might be difficult to perform the complete ILS monitoring method as described including the refillable C1 on all NDACC sites (some sites are remotely operated, others depend on support by local staff for operation during most of the time), but we feel that the sole application of the current scheme does not assure network-wide consistency of observed atmospheric trace gas columns down to the sub-percent level. We plan to perform further comparisons of ILS results derived from pressure-monitored N_2O cells with results derived from the NDACC HBr cells to which we have

access. Because the HBr cells have a comparatively high filling pressure, this comparison might reveal moderate systematic uncertainties of the HBr cell approach due to variations of the total cell pressure between different batches of cells, or due to the uncertainty of the HBr line-broadening parameters.

The final network procedure might be to define a subset of ILS reference sites within the NDACC where the complete proposed scheme is performed regularly and to send sets of sealed cells of the kind C2 and C3 back and forth between these reference sites and the other NDACC spectrometers, which for the first time will allow for a direct check of the calibration consistency between different NDACC spectrometers on the sub-percent column level, with respect to both narrow (“stratospheric”) and pressure-broadened (“tropospheric”) spectral lines. If a tested site is not able to reproduce the columns of the calibration cells based on the ILS derived from local HBr measurements, then further investigations will be required.

Finally, we plan to investigate suitable cell parameters to achieve a similar extension of the current TCCON ILS determination scheme including a closure experiment which is applicable for NIR measurements.

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