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Sensitivity of instrumental line shape with respect to different optical attenuators and resulting error propagation into atmospheric trace gas retrievals

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Abstract: The TCCON (Total Carbon Column Observing Network) and most NDACC (Network for Detection of Atmospheric Composition Change) assume an ideal ILS (Instrumental line shape) in spectra retrieval and insert an attenuator or select a smaller entrance aperture to take some intensities away if incident radiation is too strong. These processes may alter the alignment of a high resolution FTIR (Fourier transform infrared) spectrometer and may result in biases due to ILS drift. In this paper, we first investigated the sensitivity of ILS monitoring with respect to various typical optical attenuators for ground-based high resolution FTIR spectrometers within the TCCON and NDACC networks. Both lamp and sun cell measurements were conducted in this analysis via the insertion of five different attenuators before and behind the interferometer. We compared the HCl profile retrievals using an ideal ILS with those using an actual measured ILS. The results showed that the total column amounts were under-estimated by about 0.4% if the ME (modulation efficiency) amplitude deviates by about -3.5%. Furthermore, the retrieval errors increased and the obvious profile deviations are shown in a height range with a high retrieval sensitivity. ILSs deduced from all scenarios of lamp cell measurements

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were compared, and were further used to derive the HCl profile from the same spectrum. As a result, the disturbances to the ILS of a high resolution FTIR spectrometer with respect to inserting different attenuators before and behind the interferometer are quantified. The resulting ILS errors propagation into gas retrievals were also analyzed. In conclusion, the alignment of the optical parts before the interferometer is more critical than those behind the interferometer, and the entrance aperture (the focus of the entrance parabolic/spherical mirror) exhibited the most critical influence. An optimum method to adapt the incident intensity of a detector was finally deduced.

Key words: TCCON, NDACC, FTIR, Instrumental Line Shape

1 Introduction

High resolution direct solar Fourier transform infrared (FTIR) spectrometry is the most precise ground-based remote sensing technique used in deriving the column-average abundance of greenhouse gases (GHGs) or mixing ratio profiles of many other trace gases (Wunch et al., 2010 and 2011; Washenfelder, 2006; Messerschmidt et al., 2010; Kurylo, 1991; Davis et al.,2001). Currently, both the two well-established operational observation networks, i.e., NDACC (Network for Detection of Atmospheric Composition Change, http://www.ndacc.org/) and TCCON (Total Carbon Column Observing Network, http://www.tccon.caltech.edu/), use high resolution FTIR spectrometers to record direct sun spectra. Both networks have operated internationally more than ten years and their results are widely used in atmospheric physics and chemistry (Angelbratt et al., 2011; Wunch et al., 2010 and 2011; Washenfelder, 2006; Messerschmidt et al., 2010). The NDACC started operation in 1991 and mainly works in mid-infrared (MIR) spectral range of 750 to 4200 cm^{-1} (Kurylo, 1991; Schneider, et al.,2008; Kohlhepp et al., 2011; Hannigan et al., 2009; Wang et al., 2015). These spectra are used to retrieve time series of mixing ratio profiles for O_3, HNO_3, HCl, HF, CO, N_2O, CH_4, HCN, C_2H_6, $ClONO_2$ (Schneider et al., 2008; Sussmann, et al., 2011) and other gases, e.g., H_2O (Notholt, et al.,1994; Palm et al., 2010), H_2O/HDO ratio (Schneider et al, 2006), OCS (Notholt, et

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al., 2003; Wang, et al., 2016) or NH_3 (Dammers et al., 2015). In contrast, the TCCON started operation in 2004 and provides highly precise and accurate column-average abundance for CO_2, CH_4, N_2O, HF, CO, H_2O and HDO derived from near infrared (NIR) spectra (Hase et al., 2012 and 2013; Wunch et al., 2011; Keppel-Aleks, et al., 2011; Warneke, et al., 2010). In order to achieve consistent results between different FTIR sites, the TCCON and NDACC have developed strict data acquisition and retrieval methods for minimizing the site to site differences (Hase et al., 2012; http://www.ndacc.org/; http://www.tccon.caltech.edu/). Interferograms are acquired with similar instruments operated with common detectors, acquisition electronics and/or optical filters. Typically, NDACC and TCCON FTIR spectrometers are the high-resolution 125HR, 120HR, 125M or 120M manufactured by Bruker Optics, Germany[†] (http://www.ndacc.org/; http://www.tccon.caltech.edu/; https://www. bruker.com/). These interferograms were first converted to spectra and later to retrieved products using dedicated processing algorithms, i.e., GFIT, PROFFIT or SFIT (Hase et al., 2006; Dohe et al., 2013; Griffith, et al., 2010). However, biases between sites may arise due to the behavior of individual spectrometers, if not properly characterized. Some of these differences result from a misalignment of an interferometer, which can change abruptly as a consequence of operator intervention or drift slowly due to mechanical degradation over time (Hase et al., 2012; Miller et al., 2007; Olsen et al., 2004). This misalignment effects can be diagnosed by the variations of instrumental line shape(ILS) (Hase et al., 1999 and 2001). It has become a part of FTIR network practice to regularly use a low-pressure calibration gas cell (HBr or HCl) to diagnose a misalignment of the spectrometer and to improve the alignment when indicated (Wunch et al., 2010; Hase et al., 1999 and 2001). A successful alignment scheme for high resolution spectrometers was proposed about a decade ago and has become the standard alignment procedure for both TCCON and NDACC (Hase et al.,2001). As a result, the individual systematic errors and site to site biases caused by misalignment due to mechanical degradation were already

† NOTE: In the TCCON network, only the Bruker HR series instruments are used. In the NDACC network, other instruments are used as well, e.g. the Bruker M series, a BOMEM in Toronto, Canada and a self built in Pasadena, USA.

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minimized.

Typically, alignment procedure is performed by using a specific optical scenario, e.g., a specified source, aperture, beam splitter, filter and detector (Hase et al.,2001). The TCCON and most NDACC assume an ideal ILS in spectra retrieval. In order to adapt the intensity of the incident radiation, an attenuator is inserted in the light path (TCCON network[‡]) or a different entrance aperture is selected. These processes may alter the specified alignment of a high resolution FTIR spectrometer and result in biases due to ILS drift. An alternative way which does not alter the alignment of a spectrometer is selecting a suitable amplifier gain depending on incoming intensities. However, this method's contribution is limited and may be plagued with deteriorating SNR (signal to noise ratio) of the spectrum. Currently, the degree of ILS changes caused by the above processes and resulting error propagation into spectra retrievals are still not fully quantified. Furthermore, if the actual ILS is left undetermined and simply assumed to be perfect, a substantial systematic error might be introduced in the retrieval. In this paper, we designed experiments to investigate the sensitivity of ILS monitoring for ground-based high resolution FTIR spectrometer with respect to different optical attenuators. This paper also analyzed how ILS changes affect the gas retrievals, due to the insertion of various attenuators. Resultantly, we propose the optimum optical attenuation method. First, we describe the experiments design and procedures performed for this study in detail. Secondly, we present the optical background removal for both lamp and sun cell measurements. Thirdly, we analyzed the propagation of an error in the ILS into TCCON and NDACC retrieval. Furthermore, we compared the ILSs deduced from all scenarios of lamp cell measurements, and used them to derive HCl profile from the same spectrum. As a result, the disturbances of different attenuators to the ILS of a high resolution FTIR spectrometer and resulting errors propagation into gas retrievals are quantified. Following the uncertainty's estimation and discussion, this paper closes with a summary.

[‡] The attenuation is left constant and is the same for all instruments because the aperture has strong influence on the instrumental line shape.

2 Experimental design

All experiments were performed with a Bruker FTS 125HR located in Bremen, Germany. This instrument is operated by Institute of Environmental Physics (IUP), University of Bremen, Germany, and it is part of the networks NDACC and TCCON since 2004 (Messerschmidt et al., 2010). The instrument's alignment was regularly checked with the 1mm entrance aperture, CaF_2 or KBr beam splitter, InGaAs or InSb detector using the HCl and HBr gas cells, respectively. The functional layout of the experimental setup is shown in Fig. 1(a).

Five different kinds of attenuators (Fig. 1(b)) were used in the experiments. Attenuator #1 is a flat metal perforated on a regular grid. Attenuator #2 restricts the diameter of a beam. Attenuator #3 blocks the opposite 1/4 pairs of a beam and lets the rest 1/4 pairs pass through. Attenuator #4 blocks half of a beam. Attenuator #5 is a 0.8 mm or 1.2mm aperture located in the entrance aperture wheel. We performed two groups of experiments within three weeks. The alignment during these experiments was not changed and was assumed to be constant. This was backed by experience, which showed that, ILS changes only slowly if ambient conditions are stable. The group one experiments inserted an attenuator behind the interferometer, i.e., in the detector compartment. The group two experiments inserted an attenuator before the interferometer, i.e., in the source compartment (see Fig.1(a)). Each group experiment was made up of 8 individual measurement scenarios. For each individual scenario, we inserted only one attenuator and selected either the internal lamp or external sun as the light source. The internal lamps for NDACC and TCCON ILS monitoring are Globar and Tungsten, respectively. All sun cell measurements were performed within one day with a clear sky condition suitable for observations. The KBr beam splitter, a filter with CWN (center wave number) of 2300cm^{-1} and FWHM (Full Width at Half Maximum) of 500cm^{-1}, and the InSb detector were selected in NDACC ILS monitoring. While the TCCON ILS monitoring used the CaF_2 beam splitter and the InGaAs detector. In group one experiment, attenuator #1~4 was inserted to a specified place just before the exit parabolic mirror. In group two experiment, attenuator #1~4

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was inserted to a specified place between the entrance parabolic/spherical mirror and its focus (i.e., the selected entrance aperture). While attenuator #5 , i.e. an entrance aperture different from the default 1mm aperture, is right in the focal plane of the entrance parabolic/spherical mirror (see Fig.1(a)). In this manner, the attenuator #5 is in the image of the light source for both TCCON and NDACC. The group one and group two experiments for TCCON were in the parallel beam and in the divergent beam, respectively. While both experiments for NDACC were in the divergent beam (see Fig.1(a)). NDACC ILS monitoring uses a HBr cell of 2cm length filled with 2 mbar of HBr. TCCON ILS monitoring uses 10 cm long cells filled with 5 mbar of HCl. The HBr and HCl cells were provided by the National Center for Atmospheric Research (NCAR, Boulder, Colorado, USA) and Caltech (Washenfelder, 2006), respectively. Both of them were calibrated by Hase (Hase et al., 2013) at the KIT Karlsruhe, Germany. We performed 60 and 6 times of repeat measurement for each TCCON ILS monitoring scenario using the internal lamp and the sun, respectively. The repeat measurement for NDACC were set at 50 and 4 times, respectively. The repeated times in sun ILS monitoring, were much less than the lamp ILS monitoring, because, non-negligible error would have arisen if the atmosphere was assumed to be stable for a longer period.

The LINEFIT software is used for the ILS calculation. It retrieves a complex modulation efficiency (ME) as a function of optical path difference (OPD), which is represented by a ME amplitude and a ME phase error (Hase et al., 1999). The ME amplitude is referred to the width of the ILS while the ME phase error quantifies the degree of ILS asymmetry. If the spectrometer meets the ideal nominal ILS characteristics, the ME amplitude would be unity, and the ME phase error would be zero along the whole interferogram. Further details are provided by Hase (2012). For comparison, the standard micro window (WM) mode rather than broadband mode were used for all ILSs retrieval and all spectra were normalized to the same level before analysis. The WM mode facilitates the background removal especially in sun cell measurements.

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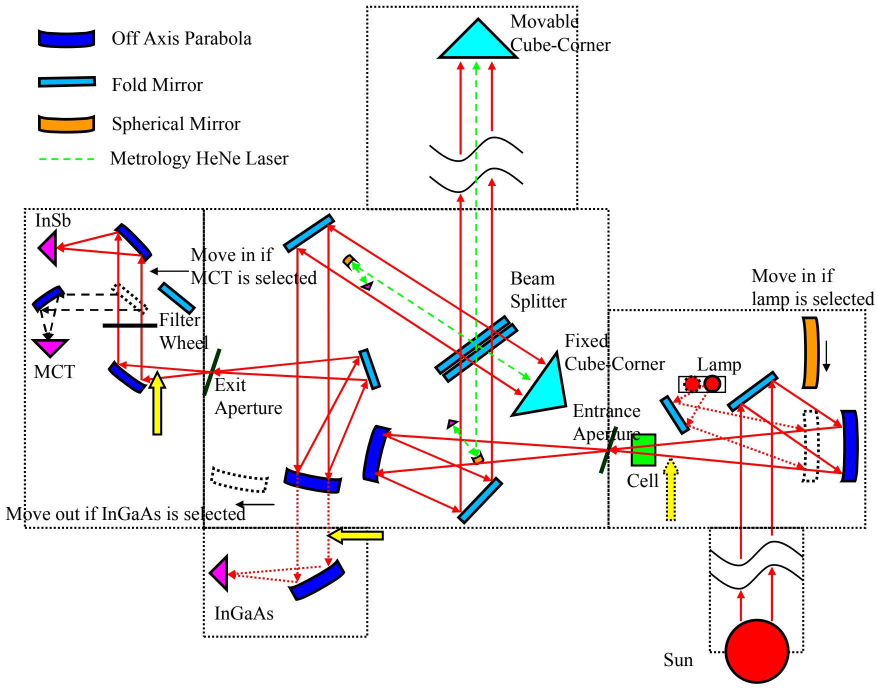
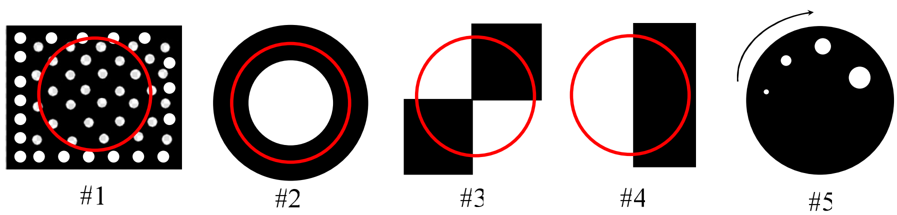


(a)



(b)

Fig. 1. Functional layout of the experimental setup, (a) the sketch of the whole optical path and (b) five different attenuators. The yellow arrows in (a) show the place where the attenuators #1~4 are inserted. In detail: the solid yellow arrows are for the classical group one experiments, i.e., attenuator #1~4 was inserted to a specified place just before the exit parabolic mirror. The dotted yellow arrow is for the group two experiments, i.e., attenuator #1~4 was inserted to a specified place between the entrance parabolic/spherical mirror and its focus (i.e., the selected entrance aperture). While attenuator #5 is right in the focal plane of the entrance parabolic/spherical mirror. The red circle in (b) indicates the size of the beam. Check the text for the detailed descriptions of the five attenuators.

3 The optical background removal

Optical background is a measurement without cells inserted into the optical path. The optical background consists of two parts. One is called the atmosphere structure, which is caused by the absorption of co-interfering gases and extinction of some atmospheric constituents. The other part is called system structure which is caused by the instrumental system, e.g., the light source, the filters, etc. In Fig.2 the typical interfering gases and the solar Fraunhofer lines within the HCl and HBr fitting regions are shown. In the TCCON case, H_2O and CH_4 have non-negligible absorptions in the same region as HCl. The NDACC case is more complicated, both N_2O and SO_2 show strong interferences in HBr region. Furthermore, non-negligible solar Fraunhofer lines within both HCl and HBr regions are shown. Therefore, optical background removal is rather important especially for the sun cell measurement. In principle, two methods can be used to remove the optical background. The first method is including all the interfering items in each fitting micro window and fit them together with the ILS. The second method is utilizing each spectrum (taken during inserting the cells) to divide by the reference spectrum (i.e., spectrum taken without cells inserted into the optical path). Since the interfering items in the solar spectrum are not easy to quantify, in this study, we used the second method to remove the optical background for both lamp and sun cell measurements.

We used the default optical settings scenario, i.e., "select the 1mm aperture without using any attenuators", to demonstrate the optical background removal for both lamp and sun cell measurements. Typical lamp and sun spectra used for TCCON and NDACC ILS retrievals are shown in Fig. 3. The sun cell measurements in both HCl and HBr regions exhibited more interference than the lamp cell measurements. The lamp measurements are nearly free of interference except the continuum curvature, whereas the atmospheric structures are obviously shown in sun measurements. This is attributable to the following reasons: a), the atmospheric measurement itself remains disturbed even a typical clear sky atmospheric condition is selected. b), the internal lamp spectra are nearly free of spectral structures, whereas the sun spectra are complex, the non-negligible Fraunhofer lines exist (as shown in Fig. 2). c), the optical path in sun cell measurement is much longer than the lamp cell measurement. The solar beam enforces the use of the interfering gases, resulting in more complicated interferences.

Fig.4 shows the fitted cases for TCCON and NDACC ILS retrievals after removing the optical background. The LINEFIT achieved good ILS fittings for both TCCON and NDACC regardless of lamp or solar spectrum. The ILS modulation efficiencies and phase errors deduced from Fig. 4 are shown in Fig.5. It concludes that the lamp

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and solar spectrum can achieve consistent ILS retrievals for both TCCON and NDACC, though the solar spectrum is much more structured than the lamp spectrum. We also performed the similar comparison for all other individual measurement scenarios, and deduced the same conclusion.

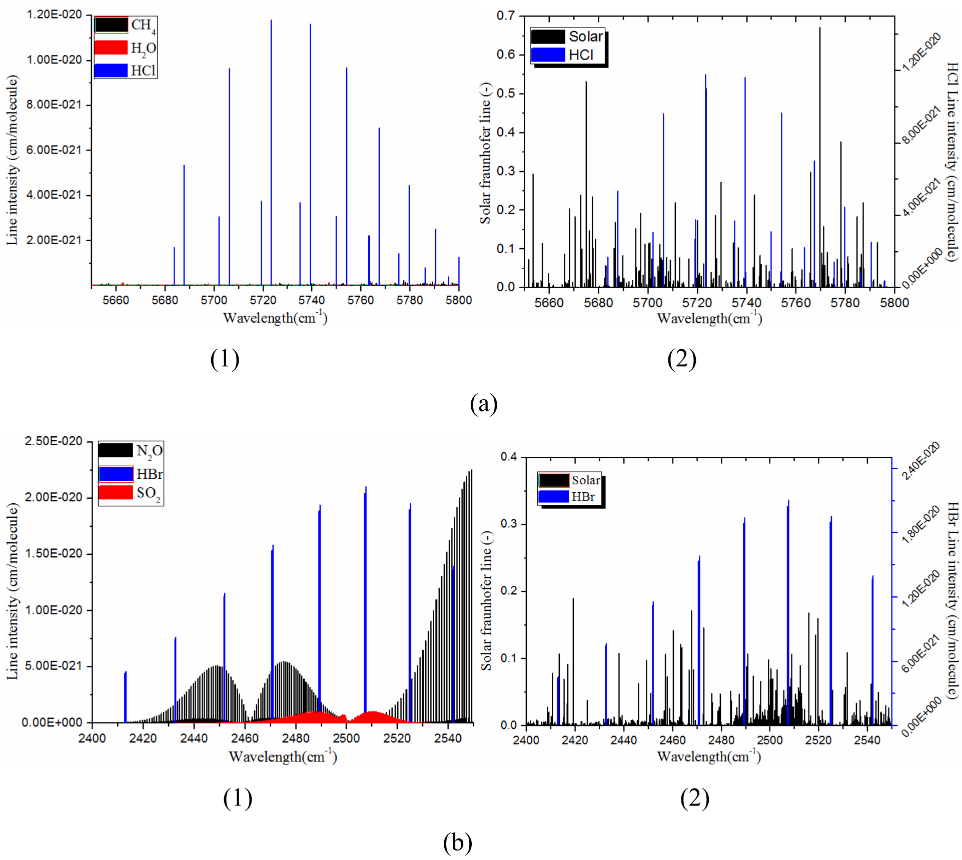

Fig. 2. Typical interfering gases and the solar Fraunhofer lines within the HCl and HBr fitting regions. (a) and (b) are the cases for HCl and HBr region, respectively. (1) and (2) of each sub panel show the interfering gases and the solar Fraunhofer lines, respectively. The absorption intensities of all gases are adopted from HITRAN2008. The solar Fraunhofer lines are adopted from the input files of the TCCON software GGG2014.

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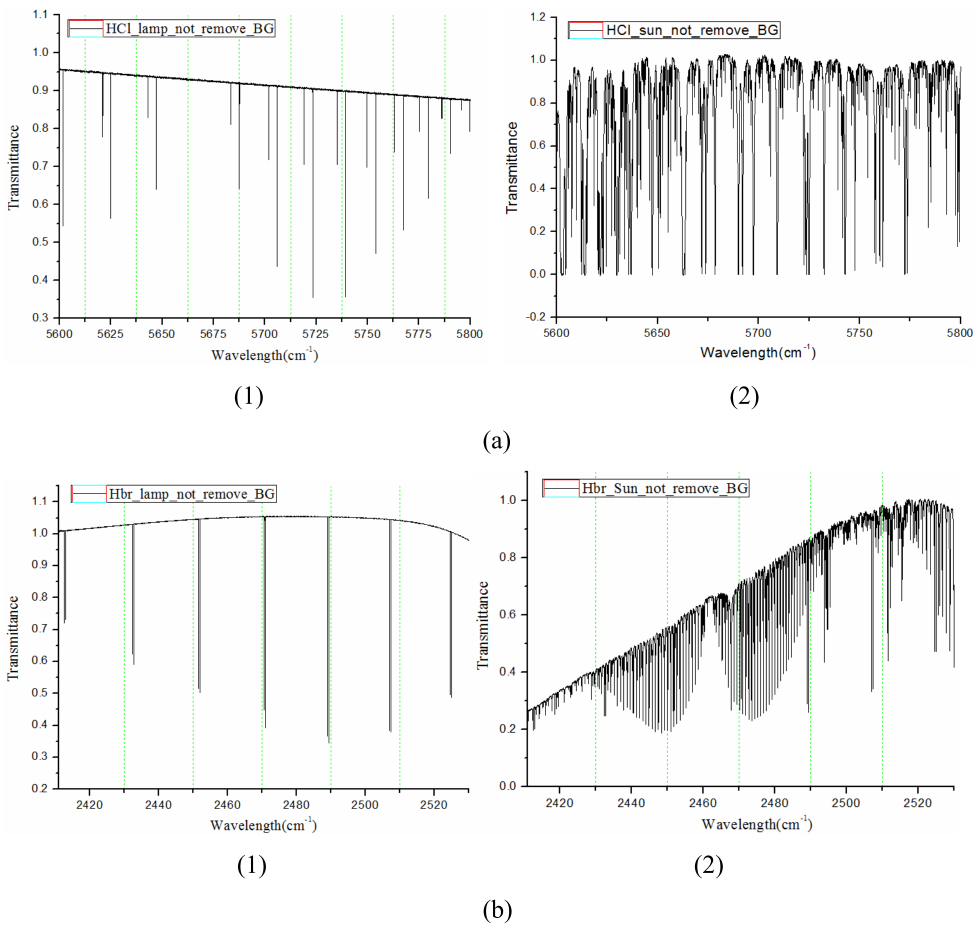

Fig. 3. The normalized spectra used for TCCON and NDACC ILS retrievals. (a) and (b) are for TCCON and NDACC ILS retrievals, respectively. (1) and (2) of each sub-plot represent lamp spectrum and solar spectrum, respectively. Less interfering structures in lamp spectra than solar spectra are shown.

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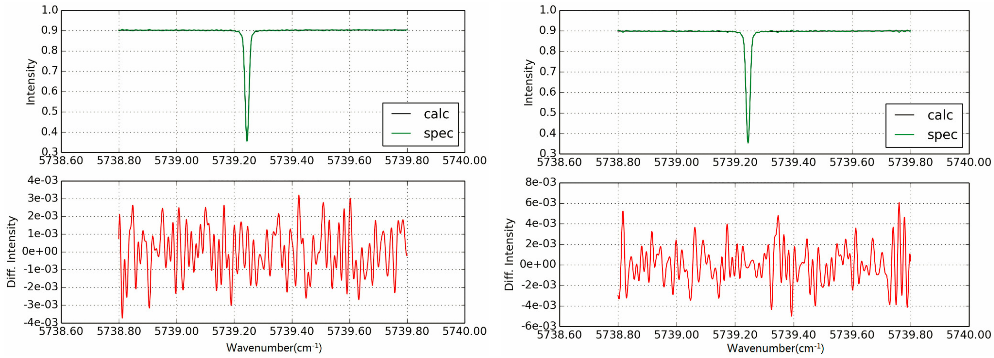
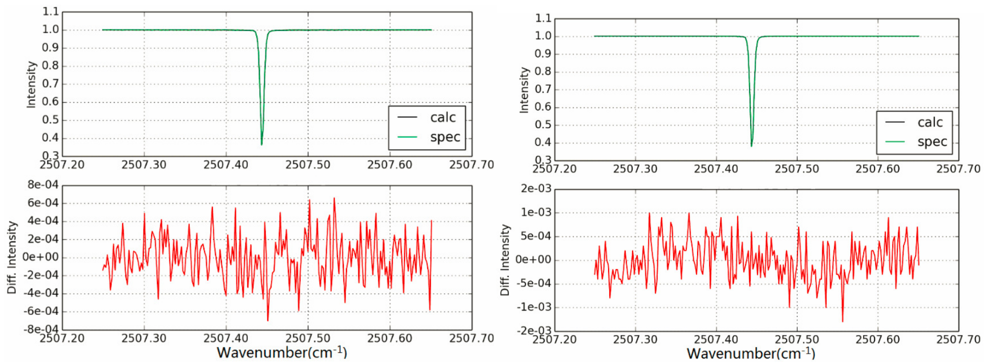


(1) (2)

(a)



(1) (2)

(b)

Fig. 4. The LINEFIT fitted cases for TCCON and NDACC after removing optical background. (a) and (b) are for TCCON and NDACC ILS fittings, respectively. (1) and (2) of each sub-plot represent the cases for lamp spectrum and solar spectrum, respectively. Only one micro-window for each case is shown and the residual are in most cases less than 0.2%.

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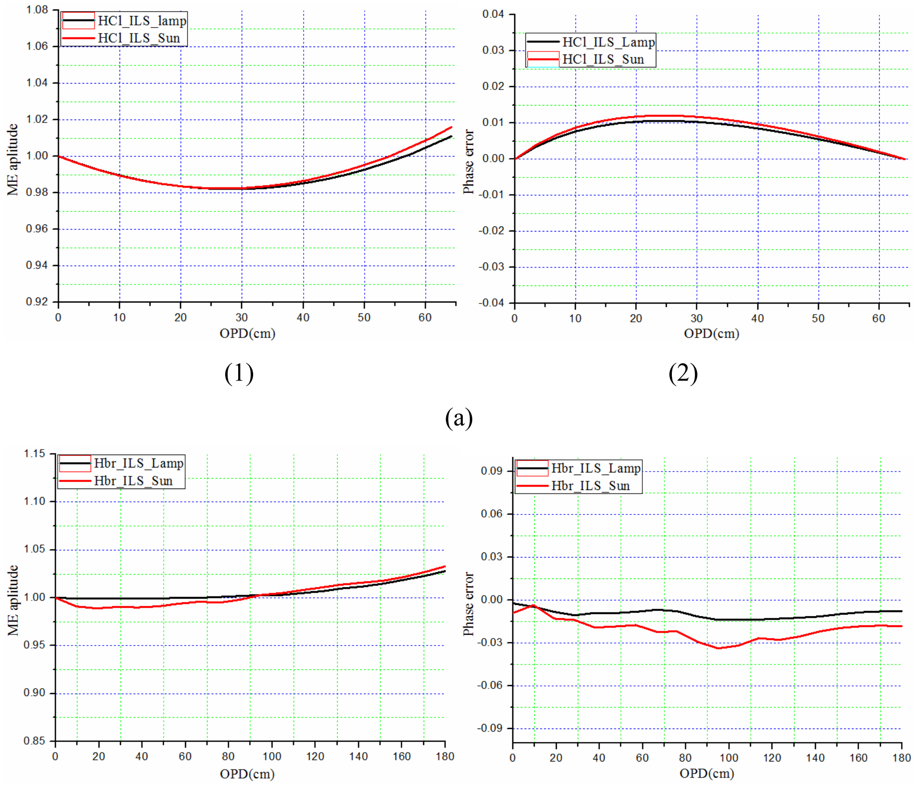

(1) (2)

(a)

(1) (2)

(b)

Fig. 5. ILS retrievals derived from lamp (black lines) and sun spectra (red lines). (a) and (b) are comparisons for TCCON and NDACC cases, respectively. (1) and (2) of each sub-plot represent the ME amplitude and phase error, respectively.

4 Propagation of ILS error into gas retrieval

Hase *et al* (2013) investigated how the column-average dry-air mole fractions of CO_2 (XCO_2) reported by TCCON are affected by a deviation of the ME amplitude from unity. They applied a disturbance on the ME amplitude and didn't take the phase error/ILS asymmetry into account due to the quantification of spectral line asymmetry was not critically affected by the assumed width of the HCl signatures provided by the cell. As a result, the propagation of ILS width error into TCCON XCO_2 data was quantified. A ME amplitude change of 4% at OPD_{max}=45cm results in XCO_2 error on the order of 0.035%. The target site-to-site bias for the TCCON XCO_2 product is 0.1%.

Therefore, the ILS error would be of secondary importance if the ME amplitude is kept at a level of less than 4% in accordance with TCCON requirements. We applied this empirical ILS error propagation formula for TCCON in this study.

The TCCON XCO_2 result is calculated from the ratio of CO_2 and O_2 columns derived from the same spectrum. This strategy minimizes the error propagation of various instrumental and model errors into the final retrieval because instrumental distortion affects the retrievals of both gases in the same manner and therefore cancel out partially (Hase et al., 2012 and 2013; Wunch et al., 2015) . The NDACC uses a different retrieval strategy, thus Hase's empirical ILS error propagation formula doesn't apply to NDACC profile retrieval. We investigated the sensitivity of NDACC data with respect to an error in the ILS by substituting the actual measured ILS derived from lamp cell measurement into atmospheric HCl retrieval. The HCl profile and column retrievals were performed using the algorithm SFIT4, version 0.9.4.4, jointly developed at the NCAR, Boulder, Colorado, USA, University of Bremen, Germany, University of Toronto, Canada, the Belgian Institute for Space Aeronomy (BIRA-IASB), Brussels, Belgium and others. The basic principle of SFIT4 is using an optimal estimation (OE) technique for fitting calculated-to-observed spectra (Rogers, 2000, Hannigan and Coffey, 2009). We applied three micro-windows, i.e., 2727cm^{-1}, 2775cm^{-1} and 2925 cm^{-1}, to retrieve HCl. Retrieval results between using an ideal nominal ILS and using an actual measured ILS were compared. In this study, we did not take the errors produced by SFIT4 into account because all the inputs were exactly the same except for ILS. The errors would be the same for all scenarios. The measured ILS and its ME amplitude and phase error along a function of OPD is shown in Fig.6. The ME amplitude deviation within 180cm OPD amounts to about -3.5%. Five spectra with different zenith angles taken from 2014/03/10 to 2014/03/20 are involved in the comparison.

The comparison with respect to total column amount, total random error and total systematic error are listed in Table 1. The results show that the total column amounts were under-estimated by about 0.4% if an ideal ILS rather than an actual ILS was used. Furthermore, an error in ILS also increased both the random error and systematic error. The random error and systematic error were increased by an order of 0.3% to 1% and ~0.5% to 1.5%, respectively. An ILS error causes more influence on systematic error than on random error, this is probably because systematic structure is made more obvious by ILS, and a correct ILS can decrease the systematic error in the forward model calculation. The comparison with respect to HCl profile retrievals is shown in Fig.7. The retrieved HCl profiles exhibit some difference when using different ILSs and the maximum deviations lie in between 20 to 60 km. In Fig.8 the retrieval errors and averaging kernels (AK) along a function of altitude for a typical HCl profile retrieval are shown. The height range where the error statistics show more retrieval errors (Fig.8 (a)) and the averaging kernels (Fig.8 (b) and (c)) show higher retrieval sensitivities are identical to the range that the retrieved HCl profiles exhibit obvious deviations in Fig.7.

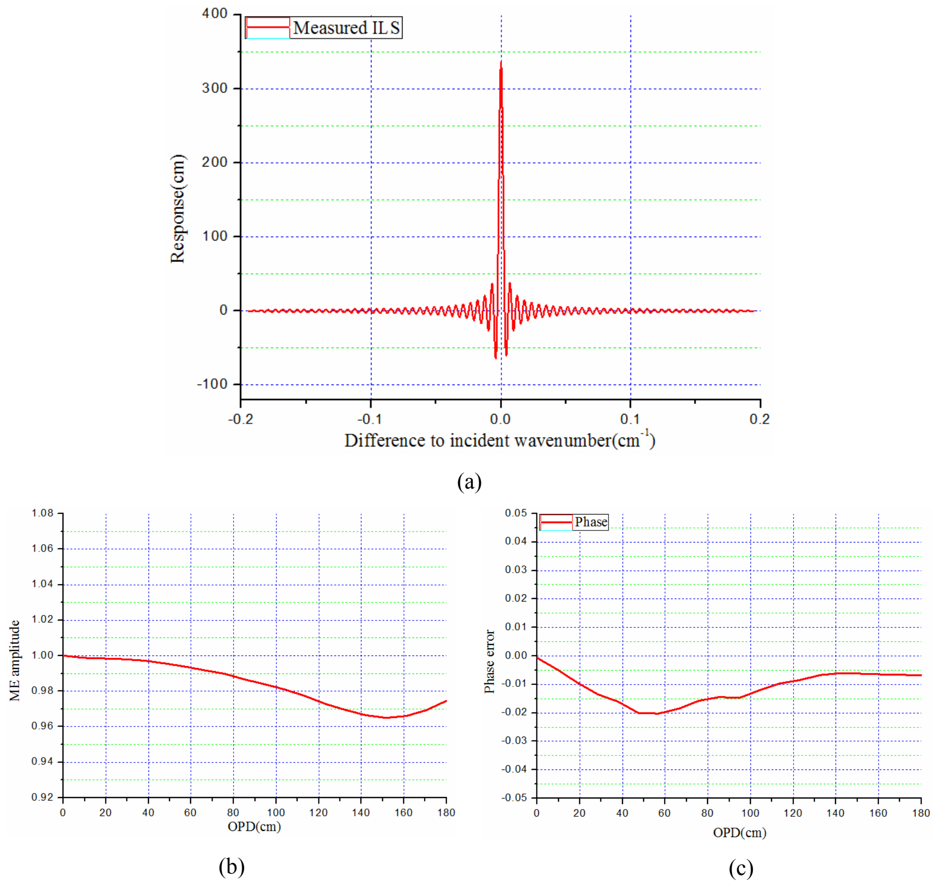

Fig. 6. The measured ILS used for HCl profile retrieval as well as its ME amplitude and phase error along a function of OPD. (a), (b) and (c) are the measured ILS, ME amplitude and phase error, respectively. The ILS was measured on 2014/11/20

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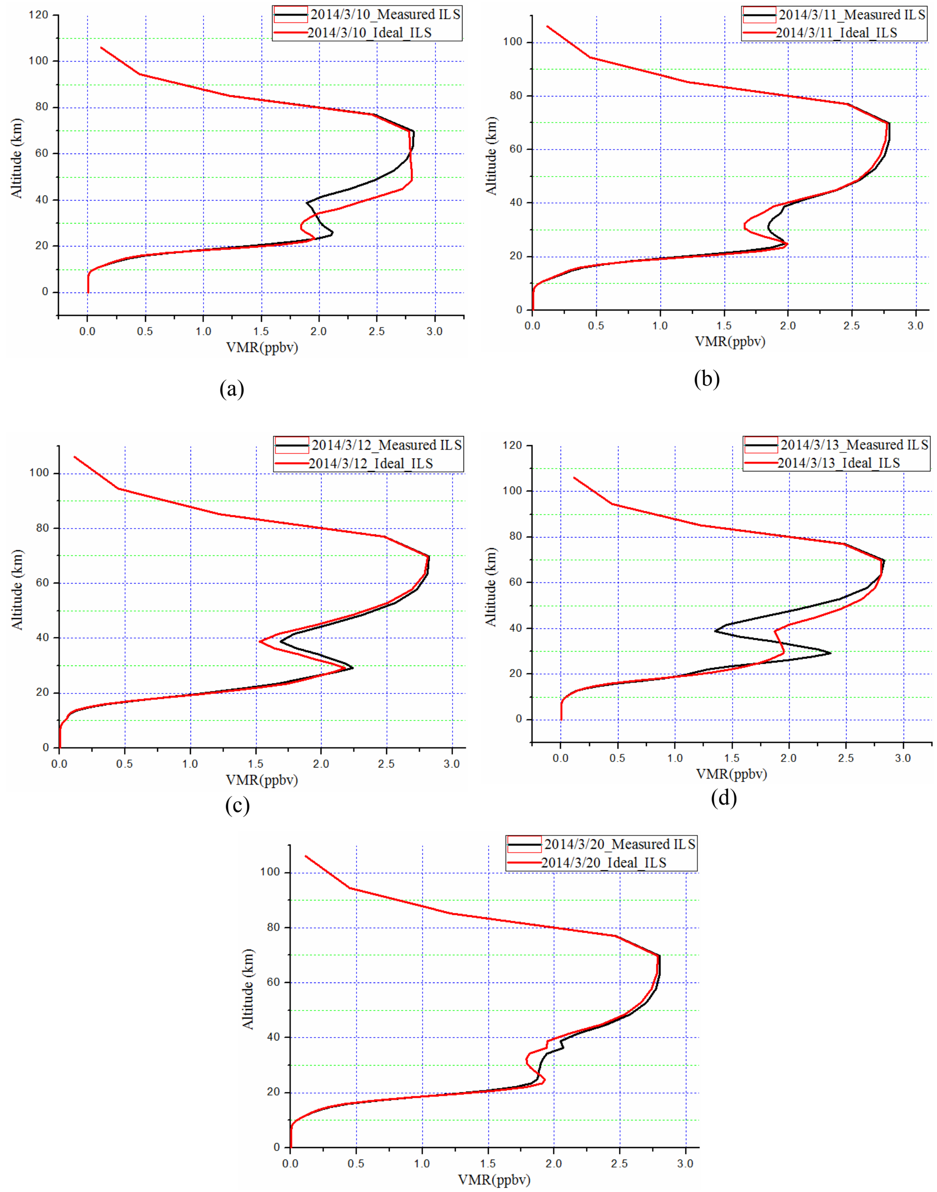

(e)

Fig. 7. HCl profile retrievals comparison between using an ideal nominal ILS and using an ILS measured on 2014/11/20. Five spectra with different zenith angles taken from 2014/03/10 to 2014/03/20 were involved. (a) to (e) are HCl profile retrievals comparison for 2014/03/10 to 2014/03/13 and 2014/03/20, respectively.

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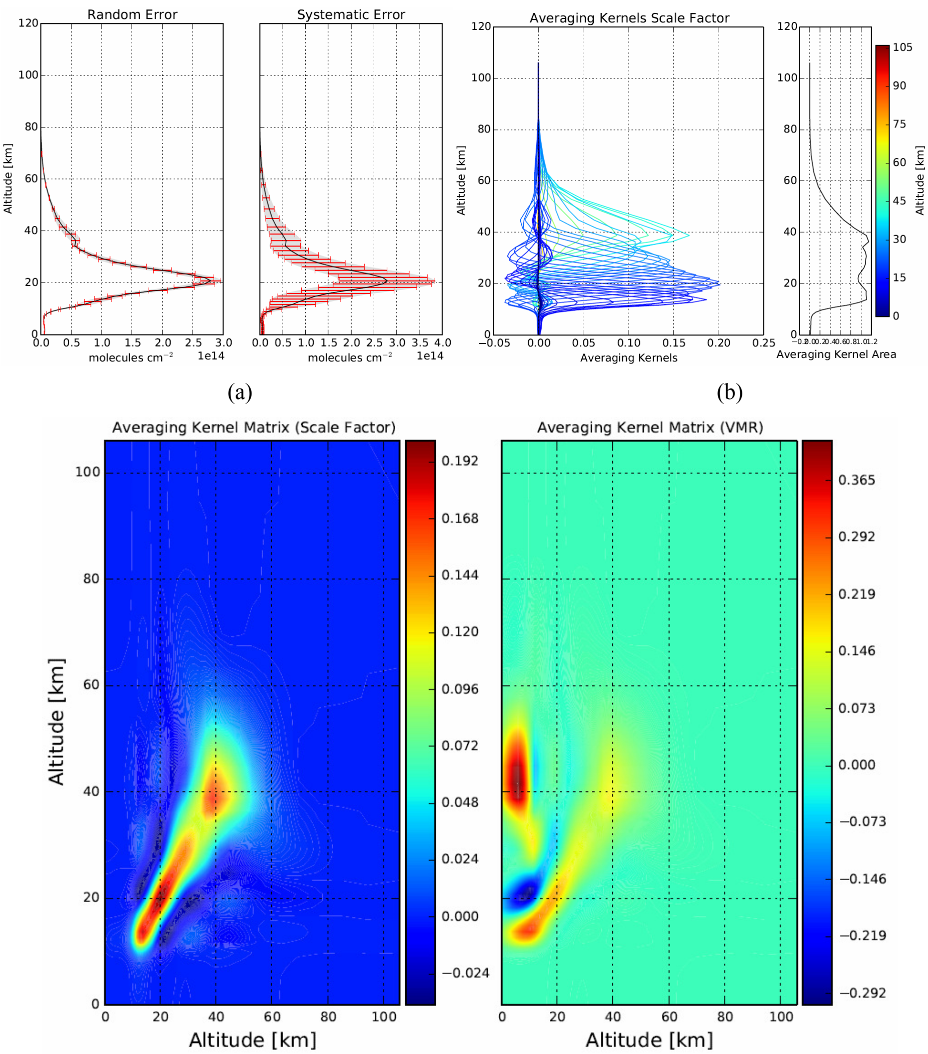

(a) (b)

(c)

Fig. 8. Retrieval error and averaging kernels(AK) of a typical HCl profile retrieval. The spectrum was taken on 2014/03/10. (a) is a plot for the retrieval error along a function of altitude. (b) and (c) are 1D and 2D plots for the averaging kernels(AK), respectively.

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Table 1 HCl retrieval results comparison between using an ideal nominal ILS and using an actual measured ILS. Five spectra with different zenith angles taken from 2014/03/10 to 2014/03/20 are involved.

Items / Date	Solar zenith angle (°)	Total column amount (E+15 molecules*cm^{-2})			Total random error (%)			Total systematic error (%)		
	Measured and ideal ILS	Measured ILS	Ideal ILS	relative bias (%)	Measured ILS	Ideal ILS	relative bias (%)	Measured ILS	Ideal ILS	relative bias (%)
2014/03/10	64.64	3.552	3.537	-0.422	0.910	0.919	+0.989	0.772	0.783	+1.425
2014/03/11	58.38	3.242	3.228	-0.432	0.979	0.985	+0.613	0.829	0.835	+1.086
2014/03/12	71.48	2.973	2.961	-0.404	1.016	1.019	+0.295	0.856	0.860	+0.467
2014/03/13	74.21	3.182	3.169	-0.409	0.874	0.880	+0.686	0.746	0.753	+0.938
2014/03/20	68.01	3.273	3.259	-0.428	0.917	0.92	+0.327	0.776	0.781	+1.03

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5 ILS retrieval sensitivity of different attenuators

The TCCON and NDACC ILS retrievals for different scenarios are presented in Fig.9 and Fig.10, respectively. The ILSs retrieved from lamp cell measurements are smoother than those retrieved from sun cell measurements for two reasons. First, we performed more times of repeat measurement for each lamp cell measurement scenario than for sun cell measurement, therefore the random noise is lower. Besides, the simpler measurement scenario makes the optical background removal of the lamp cell measurement easier and better. This is backed by Fig.4, where the fitted residual for sun spectra are relatively larger than those for lamp spectra.

It can be concluded that the ILS retrievals are very sensitive to various attenuators. The phase errors vary more than the ME amplitude. They indicate that the alignment of the interferometer was changed after either attenuator was inserted, and it caused more influence on optical modulation phase than optical modulation efficiency. The shift amounts of the ILS depends on the attenuator type. The ILS shifts caused by inserting attenuators #1~4 are much less than attenuator #5. Both TCCON and NDACC ILSs derived from cell measurements with inserting attenuators #1~4 are close to the ILS derived form default cell measurement scenario, with a ME amplitude change of < 3% within OPD_{max}=45cm and < 6% within OPD_{max}=180cm, respectively. While both TCCON and NDACC ILSs derived from cell measurements with attenuators #5 are larger than 15% at the OPD_{max}. This is most likely because the routine alignment procedure was performed by using a specified 1 mm entrance aperture, and the consistency between different apertures produce a non-negligible optical misalignment if a different aperture other than 1mm aperture was selected. This is because of mechanical inaccuracies in the mechanics of the front aperture. As the ILS asymmetry is less critical than ILS width, we set ME amplitude changes of 4% within OPD_{max} = 45cm and 8% within OPD_{max} = 180cm as the upper thresholds for TCCON and NDACC, respectively. They amount to maximum gas retrieval biases related to ILS drift on the order of ~0.035% and ~ 0.8% for TCCON XCO_2 and NDACC HCl, respectively. As a result, either of the attenuators #1~4 could potentially taken to decrease the intensity when the incident radiation is too strong. Furthermore, we also verified some derivatives of attenuators #1~4 as shown in Fig.11 which are also potential solutions. While selecting a smaller (bigger) entrance aperture to decrease (increase) incoming intensities which is less than optimal since the mechanical errors of different apertures may be non-negligible and inconsistent. This may be different from one instrument to the other, hence, the mechanical consistency of each aperture is recommended to be further checked before being used. The attenuator #5 has more influence on ILS than attenuators #1 ~ 4, which also indicates that the alignment of the entrance aperture (the focus of the entrance parabolic mirror) is more critical than other optical places.

In order to find an optimum choice from attenuators #1 ~ 4, especially for the case that the amplifier gain of a detector can not be decreased any further, we investigated the sensitivity of NDACC data with respect to these four attenuators. ILSs derived from two groups of lamp cell measurements using attenuators #1 ~ 4 were used to retrieve atmospheric HCl, and compared with the results using an ideal ILS. Section 4 shows that an error in the ILS nearly produced the same bias to all spectra. Here we only take the spectrum recorded on 2014/03/10 as an example. In contrast to section 4, here the retrieval using an ideal rather than a measured ILS was taken as the reference. In this manner, the ILSs with less biases deviated from the reference, the less ME amplitude deviated from unity and phase error deviated from zero, and the more closer to an ideal ILS assumption. The HCl total column amount retrieved by using different ILSs are listed in table 2, the total random error and total systematic error are also included. The HCl profile retrievals by using different ILSs are shown in Fig.12. The results show that both the random error and systematic error were improved a little bit by using the ILSs derived from either scenario of cell measurements. The scenarios with inserting the attenuators behind the interferometer are in most cases better than those inserting the attenuators before the interferometer. This indicates the alignment before the interferometer is more critical than that behind the interferometer. This deduction is in excellent agreement with Hase's alignment scheme (Hase, 2001). All the HCl profile retrieved via ILSs derived from cell measurements showed some deviations from the result retrieved with an ideal ILS assumption. While the result for inserting the attenuator #1 behind the interferometer exhibits the least deviation. As a

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result, we conclude that inserting an attenuator #1 behind the interferometer is the best option. It can produce a scenario that is close to an ideal ILS assumption if the interferometer is already well aligned. On the other hand, it's not an easy task to precisely insert the attenuators #2 ~ 3 in the center of an optical path or attenuators #3~4 vertically block the optical path. Errors as this kind mainly depend on personal experience and may be different from time to time. All these technical errors would produce an error to ILS retrieval because the cross-section of the optical image passing through the interferometer is not symmetric and evenly distributed. To this point, attenuator #1 is also advantageous because it homogeneously attenuate intensities in all directions and does not have this problem.

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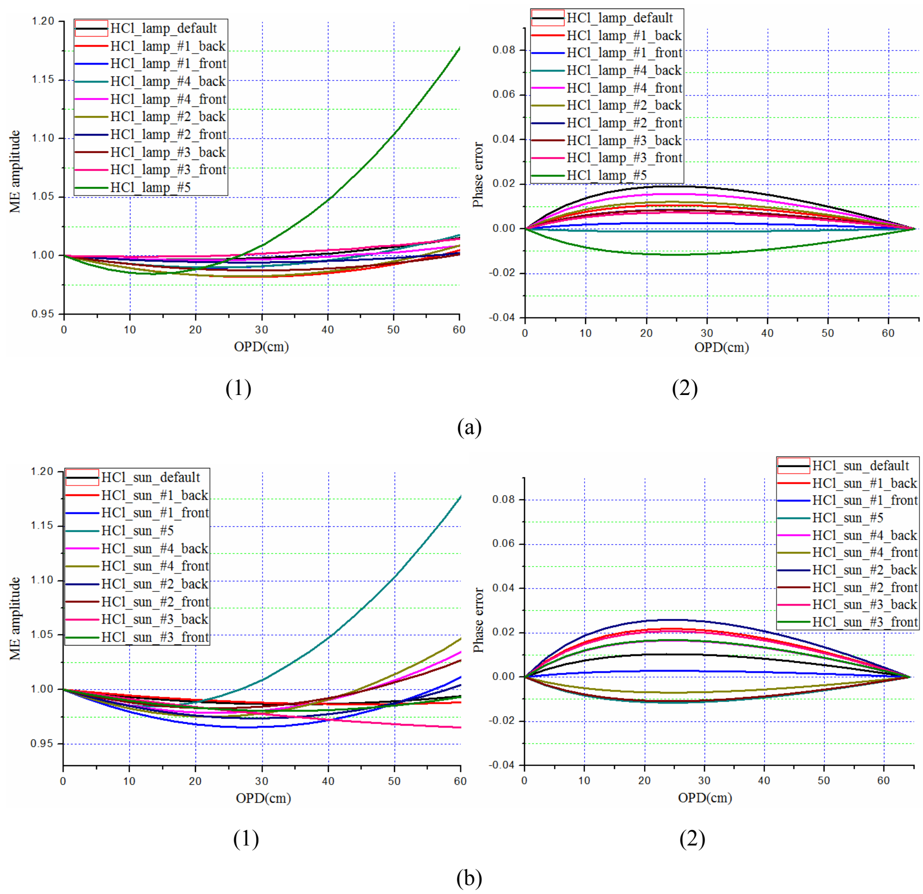

385 Fig. 9. TCCON ILS retrievals for different attenuators. (a) and (b) are ILS retrievals derived
386 from lamp and sun cell measurements, respectively. (1) and (2) of each sub-plot represent the ME
387 amplitude and phase error, respectively. The backgrounds in all scenarios were removed.
388 "HCl_sun_#3_front" represents the sun HCl cell measurement performed by inserting the
389 attenuator #3 before the interferometer. The nomenclature for other plot labels is straightforward.

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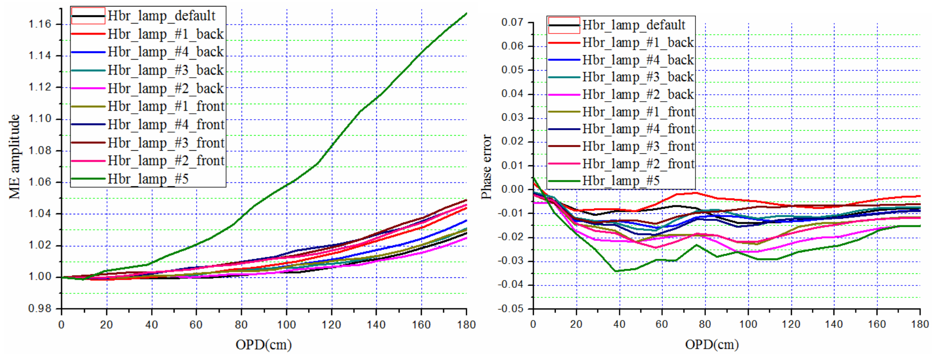
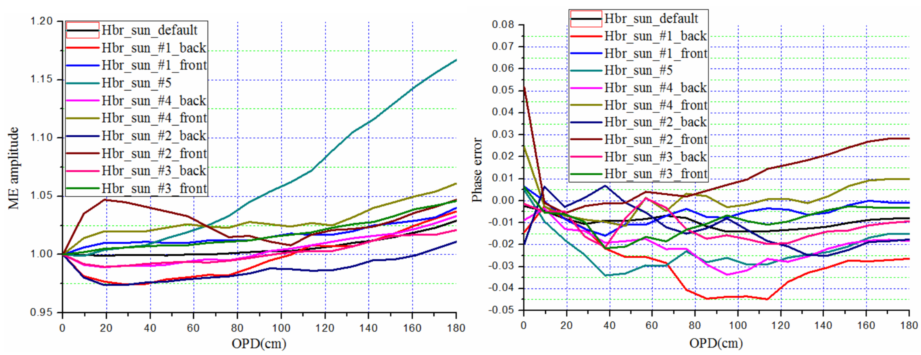


(1) (2)

(c)



(1) (2)

(d)

Fig. 10. The same as Fig.9 but for NDACC.

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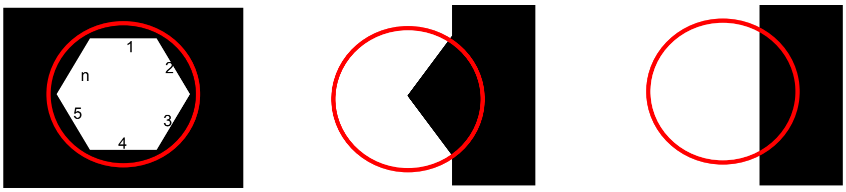

(a) (b) (c)

Fig. 11. Typical derivatives of attenuators #1~4 as potentials to decrease the incident intensity. (a) is a derivative of attenuator #2 which restricts the diameter of a beam with a Polygon. (b) is derivative of attenuator #3 which block a beam with an arc of Sector. (c) is a derivative of attenuator # 4 which partly blocks a beam.

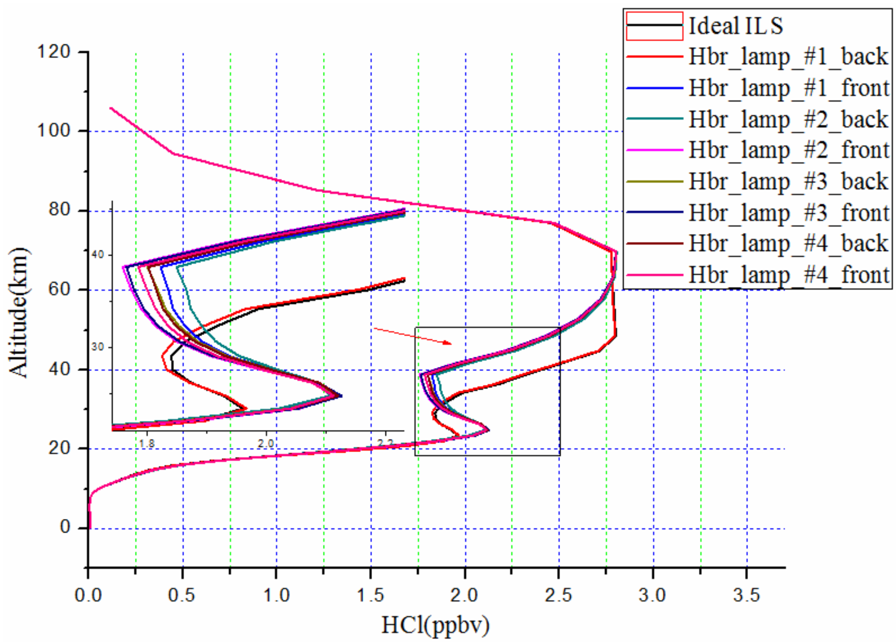

Fig.12 HCl profile retrievals comparison between using an ideal nominal ILS and using various ILSs derived from lamp cell measurements. Spectrum saved on 2014/03/10 is taken as an example. The zoom in zone is also shown. All those HCl profile retrievals by using ILSs derived from cell measurements show some deviations from the result retrieved with an ideal ILS assumption. While the result for inserting the attenuator #1 behind the interferometer exhibits the least deviation.

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Table 2 HCl retrieval results comparison between using an ideal nominal ILS and using various ILSs derived from lamp cell measurements. ILSs derived from two groups of lamp cell measurements using attenuators #1 ~ 4 were are involved. Spectrum taken on 2014/03/10 is taken as an example.

Items / Scenarios	Solar zenith angle (°)	Total column amount (E+15 molecules*cm^{-2})		Total random error (%)		Total systematic error (%)	
	–	values	relative bias (%)	values	relative bias (%)	values	relative bias (%)
Ideal ILS	64.64	3.537	0	0.919	0	0.783	0
HBr_lamp_#1_back	64.64	3.548	+0.311	0.918	-0.109	0.782	-0.128
HBr_lamp_#1_front	64.64	3.549	+0.339	0.912	-0.762	0.775	-1.022
HBr_lamp_#2_back	64.64	3.548	+0.311	0.913	-0.653	0.776	-0.894
HBr_lamp_#2_front	64.64	3.552	+0.424	0.911	-0.871	0.774	-1.149
HBr_lamp_#3_back	64.64	3.546	+0.254	0.913	-0.653	0.776	-0.894
HBr_lamp_#3_front	64.64	3.547	+0.283	0.912	-0.762	0.775	-1.022
HBr_lamp_#4_back	64.64	3.549	+0.339	0.912	-0.762	0.775	-1.022
HBr_lamp_#4_front	64.64	3.546	+0.254	0.913	-0.653	0.776	-0.894

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6 Uncertainties estimation and discussion

One uncertainty in both sun and lamp ILS measurements arises in neglecting the alignment drifts due to mechanical degradation in all experiments. This uncertainty is of secondary importance because we performed all experiments within three weeks which was much shorter than a typical realignment interval of several months. To estimate this uncertainty, we compared the ILSs before and after all the experiments. The maximum drifts of ~2.5% and ~3.6% are shown for OPD_{max} = 45cm and OPD_{max} = 180cm, respectively. They amount to the deviations of ME amplitudes of 0.0375% and 0.126% for TCCON and NDACC, respectively. We typically accomplished two individual lamp measurement scenarios per day and all solar measurement scenarios within one day. We assumed the mechanical degradation of the instrument varies evenly with the time. As a result for the lamp cell measurements, the effective ME drifts for each comparison set of TCCON and NDACC are 0.00893% (0.0375%*5/21) and 0.03% (0.126%*5/21), respectively. They amount to the maximum gas retrieval biases of ~0.0000781% and ~ 0.003% for TCCON and NDACC data, respectively. For the sun cell measurements, all estimations are ~1/5 of above deductions.

Another common uncertainty arises in errors in inputs of LINEFIT. Since we performed the same routine pro-processing procedures for all spectra and all inputs are the same except for the cell temperature. Thus, the uncertainty mainly comes from an error in a priori temperature estimation. Typically, we adjusted the a-priori temperature to find a retrieved temperature with ± 2.5K accuracy. The scatters of the effective ME drifts (normalized to a reference temperature of 296 K) are 0.0253% and 0.0591% for OPD_{max} = 45cm and OPD_{max} = 180cm, respectively (Hase, 2013). They amount to maximum gas retrieval biases on the order of ~0.0002214% and ~ 0.00591% for TCCON and NDACC data, respectively.

Assuming the atmosphere was undisturbed within the interval of each measurement scenario, would also produce uncertainties in ILS retrievals if they are derived from sun cell measurements. The clear sky condition offers relative less disturbances of aerosols, clouds and dusts, etc. Furthermore, the WM mode of

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LINEFIT makes each fitting window to be less interfered by other atmospheric molecules. So the uncertainty would mainly come from atmospheric HCl/HBr absorption variation due to SZA deviation. For a typical measurement interval of 15 minutes, a SZA deviation of ~2.5° can normally be observed at SZA = 60°. This amounts to ~8.3% ({[1/cos(62.5°)-1/cos(60°)]/(1/cos(60°))}) of atmospheric HCl/HBr absorption deviation. For the TCCON case, the typical atmospheric HCl total column amount as listed in table 1 is on the order of E+15 molecules*cm^{-2}. This means the atmospheric HCl absorption deviation is on the order of E+14 molecules*cm^{-2}. While the HCl total column amount within the HCl cell is on the order of E+22 molecules*cm^{-2} (Table 1 in Hase etal., 2013; http://www.tccon.caltech.edu/). Consequently, uncertainties as this kind are also negligible. A similar deduction for the NDACC case results in the same conclusion.

In conclusion, this study is accurate enough to resolve the ILS of each cell measurement scenario. Our study cannot identify the physical mechanisms of how the different attenuators varied the alignment of a ground-based high resolution FTIR spectrometer within the TCCON and NDACC networks, but our findings shall provide a valuable reference for all TCCON and NDACC communities because all these FTIR networks nearly operate with the same hardware and software.

7 Summary

We investigated the sensitivity of ILS monitoring for ground-based high resolution FTIR spectrometer with respect to various typical optical attenuators. We performed both lamp and sun cell measurements via the insertion of five different attenuators before and behind the interferometer to derive different ILSs. We found that both solar spectrum and the lamp spectrum can achieve consistent ILS if the optical background is properly removed. We compared the HCl profile retrievals using an ideal ILS with those using an actual measured ILS. The results showed that the total column amounts were under estimated by about 0.4% if a ME amplitude is deviated by about -3.5%. Furthermore, the retrieval errors increase and profile deviations are obviously shown in a height range with a high retrieval sensitivity. ILSs deduced from all scenarios of lamp cell measurements are compared, and are further used to derive HCl profile from the same spectrum. As a result, the influence of different attenuators on the ILS of a high resolution FTIR spectrometer are quantified. The worst option to increase (decrease) the intensity is selecting a bigger (smaller) entrance aperture. This is because the mechanical errors of different apertures may be non-negligible and inconsistent. Inserting attenuators #1 ~ 4 before or behind the interferometer can be used to adapt the intensity of a detector. While inserting a grid-like attenuator #1 behind the interferometer is the optimum option. It can produce a scenario close to an ideal ILS assumption if the interferometer is already well aligned.

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