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ground-based Brewer
and FTIR technique

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Comparison of ground-based Brewer and FTIR total O₃ monitoring techniques

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Abstract

We compare the currently most precise, ground-based total O₃ measurement techniques: Brewer and FTIR. We give an overview of the similarities and the differences between the measurements and the retrieval approaches of both experiments. By means of coinciding measurements performed at the Atmospheric Observatory of Izaña from 2005 to 2007 we show that the small differences between both techniques are in excellent agreement with the theoretical predictions. Our empirical study confirms that both FTIR and Brewer measurements are able to continuously monitor total O₃ amounts with a precision of better than 0.4%.

1 Introduction

Over the last 20 years significant progress was made concerning the precision and accuracy of atmospheric remote sensing measurements. This development is due to improvements in both instrumental setups and in retrieval algorithms. High precision measurements performed on a continuous basis are needed in order to detect potential atmospheric trends as soon as possible. A good example is the atmospheric ozone content. In the coming decades some kind of ozone recovery is expected, however, it is difficult to predict how, when, and to what extent it will occur (Weatherhead and Andersen, 2006). Continuous high precision measurements of O₃ are very important in this context. They provide for improved atmospheric O₃ models by detecting trends between the models and the measurements as soon as possible.

Currently there are two continuous ground-based measurement techniques that claim to monitor total O₃ with a precision of 1–2 DU: FTIR and Brewer. Their precision and accuracy are theoretically estimated in Schneider and Hase (2007) and Redondas and Cede (2006), respectively. Ground-based measurements are an important component of the global atmospheric monitoring system, since they are decisive for the validation of satellite data. Only space-based measurements achieve a

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continuous global coverage, but their quality is coupled to the precision and accuracy of the ground-based long-term reference experiments. In particular so-called “super-sites” play an important role in this context. They concentrate different measurements techniques at the same site offering a variety of parameters to be used for the validation of satellite data. Furthermore, “super-sites” are important for an ongoing improvement of the precision of the different techniques, since they allow a check of the theoretical error estimations under routine conditions by inter-comparing the different experiments. In this work we compare total O₃ amounts obtained from a state-of-the-art FTIR observing system and from the European reference Brewers. The measurements are performed at the Izaña Observatory, Canary Island of Tenerife, Spain. Currently the Izaña FTIR system provides world-wide the most precise FTIR O₃ data, which is due to both advanced instrumental equipment and an optimised O₃ retrieval strategy (Schneider and Hase, 2007). Concerning the Brewer, Izaña is the Regional Brewer Calibration Centre for Europe (<http://www.rbcc-e.org/>) of WMO/GAW (World Meteorological Organisation/Global Atmospheric Watch), which guarantees highest quality standards. Both FTIR and Brewer activities are part of NDACC (Network for Detection of Atmospheric Composition Change; Kurylo, 1991; 2000; <http://www.ndacc.org/>). Such high quality and continuously performed FTIR and Brewer measurements only coincide at the Izaña Observatory. Therefore, it is a predestinated site for an inter-comparison study of both techniques.

In the following we list the main similarities and differences of the FTIR and Brewer technique, and discuss their respective advantages and disadvantages (Sect. 2). In Sect. 3 we briefly introduce the site where the measurements used in this work were made. In Sect. 4 to 6 we compare operational data from two different Brewer spectrometers and the FTIR spectrometer. Finally, we summarize the reasons that make the Izaña data unique and conclude about the implications of our results (Sect. 7 and 8).

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2 FTIR versus Brewer technique

The principle of both techniques is the same: both the FTIR and the Brewer spectrometer detect direct solar light. The experimental differences are the spectral regions that are analysed and the spectral resolution of the measurements. The respective retrieval algorithms deduce the atmospheric O₃ amounts by analysing the absorption signatures imprinted onto the extraterrestrial solar spectrum. For this purpose the Brewer and FTIR technique use different retrieval approaches, which are imposed by the nature of the respective experimental data.

2.1 The experiments

The Brewer spectrometer detects spectral irradiance in six channels in the UV (303.2, 306.3, 310.1, 313.5, 316.8, and 320.1 nm) each covering a bandwidth of 0.5 nm (resolution power $\lambda/\Delta\lambda$ of around 600). The spectral analysis is achieved by a holographic grating in combination with a slit-mask which selects the channel to be analysed by a photomultiplier. At Izaña only MK-III Brewer instruments with double monochromators are applied which widely reduces the impact of straylight on the measurements. The Brewer system works in a completely automatic way, and usually measures continuously during the whole day. The first channel at 303.2 nm is only used for spectral wavelength checks by means of internal Hg-lamps, the second channel is used for measuring SO₂ and the remaining four channels at longer wavelength determine the O₃ amounts. The finally reported O₃ result is the mean value of a set of 5 observations. The standard deviation of these 5 observations is used for the acceptance of the measurement. In this work we consider a measurement to be valid if the standard deviation is lower than 2.5 DU. The Brewer's field of view (FOV) is about 2.7° (diameter of solar disc is 0.5°). Consequently, all direct solar irradiance is coupled into the spectrometer even for a moderate misalignment of the solar tracker. On the other hand, a certain fraction of the diffuse radiance (circumsolar) is measured together with the direct irradiance. This signal of the diffuse radiance increases with the amount of scattering, i.e.

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mainly with SZA and aerosols, and alters the retrieved O₃ amounts.

The Brewer experiment needs some instrumental characteristics to be determined by calibration experiments: the wavelength calibration and the slit function (instrumental line shape function), and the extraterrestrial constant (ETC). The slit function are determined once per year. This is done in a laboratory by means of low pressure discharge lamps (e.g. Fioletov et al., 2005; Gröbner et al., 1998). The exact wavelength settings are monitored in an automated mode every 40 min by means of the internal Hg lamps. The slit function and the exact wavelength settings are necessary to convolve the highly resolved Bass and Paur (1985) O₃ absorption coefficients with the instrumental function. The Izaña sky conditions allow the determination of the ETC for each Brewer independently by the Langley method. However, in this work we use the ETC transferred by the traveling world standard Brewer #017. This ETC calibration is done once per year. The traveling world standard is applied to guarantee a world-wide consistency within the Brewer network. The stability of the ETC calibration is monitored continuously in an automated mode using an internal halogen lamp. The difference between the weighed counts of the halogen lamp at the time of calibration and the smoothed mean of two weeks is used to correct the ETC (Fioletov et al., 2005). For the period of this work only minor corrections are applied (less than 0.3%), which indicates very stable instrumental characteristics.

The FTIR spectrometer measures in the middle infrared (between 1015 and 780 cm⁻¹, which corresponds to 9.8 and 12.8 μm). The spectra are obtained by a Fourier analysis of the recorded interferograms. The spectral resolution depends on the maximum optical path difference of the interfering light beams. For operational O₃ measurements the FTIR spectra are measured with a resolution of 0.005 cm⁻¹, which corresponds to a resolution power $\lambda/\Delta\lambda$ of around 2×10^5 . The FTIR measures continuous spectra over a broad wavelength range. The spectral windows applied for the O₃ retrieval consist of a total of 7500 spectral bins and contain more than 100 individual O₃ rotational-vibrational lines and 4 CO₂ lines of different intensity. The CO₂ signatures allow the retrieval of temperature profile which is expected to widely improve the

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precision of the retrieved total O₃ amount (Schneider and Hase, 2007). It is important to mention that all these individual rotational-vibrational lines are resolved: in the middle infrared the shape of absorption lines is dominated by pressure broadening with a typical HWHM of 0.04 cm⁻¹ at surface level, which is nearly one order of magnitude larger than the operational resolution of the FTIR spectrometer. The numerous fine-structured spectral features provide for an automatic wavelength calibration. The FTIR spectrometer has a field of view of only 0.2°, i.e. it only analyses sunlight coming from the center of the solar disc (diameter of 0.5°). Consequently, a misalignment of the solar tracker directly affects the observing geometry. In Schneider and Hase (2007) it is shown that for elevation angles below 20° this is the leading error source. Although on a much higher level of spectral resolution if compared to the Brewer, it is important to monitor the ILS of the FTIR spectrometer. This is done regularly by performing low pressure cell calibration measurements as described in Hase et al. (1999). Even then the residual ILS error is estimated to be the most important error source concerning FTIR total O₃ amounts measured at solar elevation angles above 20°.

2.2 The retrieval approaches

The basic equation for analysing solar absorption spectra is Lambert Beer's law:

$$I(\lambda) = I_{\text{ET}}(\lambda) \exp(-\tau_{\text{O}_3}(\lambda) - \sum_x \tau_x(\lambda)) \quad (1)$$

Here $I(\lambda)$ is the measured intensity at wavelength λ , I_{ET} the extraterrestrial intensity, τ_{O_3} the optical depth due to O₃, and τ_x the optical depth due to all other atmospheric components (other trace gases, aerosols, ...). For O₃ it is:

$$\tau_{\text{O}_3}(\lambda) = \int \sigma_{\text{O}_3}(\lambda, s) n_{\text{O}_3}(s) ds \quad (2)$$

whereby $\sigma_{\text{O}_3}(\lambda, s)$ is the absorption cross section and $n_{\text{O}_3}(s)$ the concentration of O₃ at location s . The integration is performed along the path of the direct sunlight. The cross section σ_{O_3} depends on temperature and in the infrared additionally on pressure.

The integration of n_{O_3} perpendicularly throughout the atmosphere gives the total O_3 column amount (Ω_{O_3}):

$$\Omega_{\text{O}_3} = \int n_{\text{O}_3}(z) dz \quad (3)$$

To derive Ω_{O_3} from the measurements both techniques apply different approaches.

5 2.2.1 Principles of the Brewer retrieval

The Brewer algorithm considers the absorption by O_3 and SO_2 , scattering by molecules, and extinction by aerosols. The algorithm applies an airmass factor (μ_x), according to:

$$\mu_x = \sec \left(\arcsin \left[\frac{R}{R + h_x} \sin \Theta \right] \right) \quad (4)$$

10 as ratio between the slant and the vertical total column amount. Here R , Θ , and h_x are the Earth's radius, the apparent solar zenith angle ($90^\circ - \text{elevation angle}$), and the effective altitude of the absorbing or scattering component ($h=22 \text{ km}$ for O_3 , SO_2 and Aerosols, and $h=5 \text{ km}$ for Rayleigh scattering). Equation 4 is a simplification of the real situation. It assumes that the absorbing compounds are concentrated at a single
 15 altitude h_x and it disregards that the refraction index depends on altitude. The errors produced by this assumption increase with decreasing solar elevation angles. For O_3 and an elevation angle of 5° it may exceed 4% [Bernhard et al. \(2005\)](#).

Taking the logarithm of Eq. 1 and applying Eqs. 2 and 3 yields the following relation between the intensities at channel i (I_i) and the amount of the extinction components:

$$20 \log I_i = \log I_{\text{ET},i} - \sum_x \mu_x \sigma_x \Omega_x - \mu_{\text{O}_3} \sigma_{\text{O}_3} \Omega_{\text{O}_3} \quad (5)$$

The four channels at the longer wavelength are combined ([Evans et al., 1987](#)),

$$\sum_{i=1}^4 w_i \log I_i = \text{ETC} - \sum_{i=1}^4 w_i (\sum_x \mu_x \sigma_x \Omega_x + \mu_{\text{O}_3} \sigma_{\text{O}_3} \Omega_{\text{O}_3}) \quad (6)$$

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and the weighting coefficients w_i are selected to minimise the influence of SO_2 : $w_{[1;4]} = [1.0, -0.5, -2.2, 1.7]$. This choice also widely eliminates absorption features which depend in local approximation linearly on wavelength (λ) like Rayleigh scattering and aerosol extinction, since $\sum_{i=1}^4 w_i \lambda_i \approx \sum_{i=1}^4 w_i = 0$. In Eq. 6 we replaced $\sum_{i=1}^4 w_i \log I_{\text{ET},i}$, the extraterrestrial coefficient, by ETC. With the wavelength and slit function calibration we can calculate the convolved extinction coefficients (σ_x of Eq. 6). The ETC is transferred from a reference instrument (Fioletov et al., 2005). Changes with time in the sensitivity of the instrument are reflected in changes of this extraterrestrial coefficient. Equation 6 together with 4 provides the total vertical O_3 amount. For the derivation of Eq. 5 we have to assume a constant so-called effective σ_x throughout the atmosphere. Any temperature or pressure dependence is neglected. The “operational” algorithm applies a σ_{O_3} corresponding to an effective height of O_3 of 22 km and a fixed effective temperature of the O_3 layer of -45°C . The assumption of an effective height also influences the calculation of the airmass factors (Eq. 4). These simplifications produce systematic errors. In the case of the Izaña Observatory the effective height is about the same as the one used by the ‘operational’ algorithm. The effective temperature ranges from -50°C in winter months to -45°C in summer months which gives in the worst case a systematic error of 0.4% in winter months.

2.2.2 Principles of the FTIR retrieval

The FTIR retrieval (PROFFIT, Hase et al., 2004) applies a precise radiative transfer model (KOPRA, Höpfner et al., 1998; Kuntz et al., 1998; Stiller et al., 1998). KOPRA is a line-by-line model which simulates the measured spectra. It includes a ray tracing module (Hase and Höpfner, 1999) to simulate how the solar light passes through the atmosphere. The model uses a discretised atmosphere (here we apply 41 levels between the Earth surface and the top of the atmosphere). The optical depth for each layer, enclosed between a pair of adjacent levels, is calculated by performing the integration of Eq. 2 between two levels. The applied absorption cross section σ for each

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individual line and level are parameterised according to the HITRAN spectroscopic database (Rothman et al., 2005). The parameterisation takes care of the pressure and temperature dependency of σ , i.e. it depends on the pressure and temperature actually present at the corresponding level. Summing up the τ values from the different layers leads to the simulated spectra at the location of the observer (combination of Eqs. 1 and 2). The radiative transfer model determines the changes in the spectral fluxes y for a changing state vector x . These derivatives are sampled in a Jacobian matrix \mathbf{K} :

$$\partial y = \mathbf{K} \partial x \quad (7)$$

Inverting \mathbf{K} of Eq. 7 would allow an iterative calculation of the atmospheric state from the measurement alone. However, generally the problem is under-determined, i.e. \mathbf{K} is singular. To overcome this problem an optimal estimation (OE) approach is applied (Rodgers, 2000): since the actual atmospheric state cannot be determined unambiguously from the measurement the OE approach determines the most probable state for the given measurement. The approach bases on the Bayesian theorem and consists in maximizing a total probability density function (pdf). The total pdf is the product of two pdfs: a first, describing the probabilities of the residuals of the spectral fit, and a second, describing the a-priori known probabilities of the absorbers' distributions.

This approach produces vertical O_3 concentration profiles ($n_{\text{O}_3}(z)$) for several O_3 isotopologues ($^{48}\text{O}_3$, asymmetric and symmetric $^{50}\text{O}_3$ and $^{49}\text{O}_3$) individually, since each isotopologue offers distinct absorption lines. The total column amounts are subsequently calculated by Eq. 3. In addition, the measured spectrum contains sufficient information to determine the main issues of the actual temperature profile. Actually the O_3 retrieval consists of a joint OE of O_3 , $^{50}\text{O}_3/^{48}\text{O}_3$, interfering species, and temperature. Further details about the O_3 FTIR retrieval and its error budget are given in Schneider and Hase (2007).

Applying highly-resolved infrared spectra it is easy to separate the extraterrestrial component from the fine-structured absorption signatures of atmospheric trace gases. In the infrared the solar radiation is reasonable close to a black body radiation at

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6000 K, i.e. it has a rather smooth dependence on wavelength. The presence of the few solar lines causes no significant problem, since they are well known (e.g. Hase et al., 2006) and fully resolved by the FTIR spectrometer.

The spectral windows applied for the FTIR retrieval contain many spectral bins between the fine-structured absorptions of atmospheric trace gases to construct an empirical background continuum.

2.3 Summary

Table 1 collects the principle differences of both techniques and resumes their respective advantages. The Brewer technique depends on several calibration experiments: (a) the wavelength setting and slit function calibration. The convolved absorption cross sections strongly depend on these calibrations. (b) The transfer of the extraterrestrial constant (ETC) from the world standard Brewer. The ETC is necessary to separate the extraterrestrial from the atmospheric signal. Errors in the ETC add directly to the slant column amounts (see Eq. 6), i.e. they are especially important for low slant column amounts. On the other hand, every single FTIR spectrum is self-calibrating with respect to the wavelength and intensity: (a) the FTIR spectrometer fully resolves many rotational-vibrational lines, it provides for a very accurate and automatic wavelength calibration of the measured spectra. (b) the extraterrestrial spectrum can be easily simulated and there is sufficient information to derive the overall instrumental transmittance from every single measurement. Nevertheless, for high precision measurements of absorbers with sharp signatures (stratospheric absorbers), it is important to monitor the actual ILS continuously. For elevation angles above 20° the remaining ILS uncertainties are the leading error source.

The Brewer algorithm applies the same σ_{O_3} throughout the year and throughout the whole atmosphere. It, furthermore, assumes a O_3 profile where all O_3 is concentrated in a single layer at 22 km. The FTIR retrieval, on the other hand, takes into account the actual O_3 and temperature profile and applies different σ_{O_3} s and temperatures for 41 different atmospheric levels. This is important, since σ_{O_3} depends on temperature and

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pressure. Furthermore, the FTIR retrieval uses a comprehensive ray tracing model.

The field-of-view of the Brewer instrument is rather large and a perfect tracking is less important. However, it simultaneously analyses a significant amount of diffuse light, which is an important error source for low elevation angles. The field-of-view of the FTIR is smaller than the solar disc. For low elevation angles (below 20°) the FTIR data quality depends critically on a perfectly working solar tracker.

A great advantage of the Brewer technique is that the measurements are performed with a very compact and mobile instrument and that all measurements including the calibrations are made nearly automatically. The measurements are performed continuously at many sites over the globe. For that reasons Brewer O₃ data are often used as reference in inter-comparison studies between different instruments and techniques, including space-based instruments. FTIR spectrometers only have limited mobility. They are generally installed inside a laboratory or a container. On the other hand, they are very versatile instruments measuring a great variety of atmospheric species. Concerning O₃ the FTIR measurements allow to distinguish between its different isotopologues. FTIR measurements at Izaña are typically performed three time per week.

3 The Izaña “super-site”

The Brewer and FTIR measurements are performed at the Izaña Observatory, which is located on the Canary Island of Tenerife, 300 km from the African west coast at 28°18' N, 16°29' W at 2370 m a.s.l. The Izaña Observatory is run by the Spanish Weather Service. It is a World Meteorological Organization (WMO) Global Atmospheric Watch (GAW) station of global importance, and there are many different institutes from different countries involved in its manifold measurement program: in-situ measurements of O₃, CO₂, CO, CH₄, N₂O, NO_x, SO₂, SF₆, ..., different in-situ analysers and filter radiometer to determine optical, physical and chemical properties of aerosols. In March 2001 Izaña's ECC-sonde, DOAS, Brewer and FTIR activities have been accepted by the NDACC (Network for Detection of Atmospheric Composition Change,

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formerly called NDSC: Network for Detection of Stratospheric Change (Kurylo, 1991, 2000); <http://www.ndacc.org/>).

The Brewer measurements at Izaña started in May 1991. Since November 2003 they represent the Regional Brewer Calibration Centre for Europe (<http://www.rbcc-e.org/>) of WMO/GAW (World Meteorological Organisation/Global Atmospheric Watch). This Calibration Centre is essential for a coordinated European Brewer network that is needed for both present and future consistency of ground-based total ozone observations and for validation of satellite instruments. Furthermore, it plays an important role in the development and testing of new measurement techniques for the whole Brewer network. The Brewer data are often used as reference for validating other ground- and satellite-based instruments.

The FTIR activities started in January 1999 and until 2005 a Bruker IFS 120M FTIR spectrometer was applied. Since January 2005 a Bruker IFS 125HR spectrometer is in operation. Currently the IFS 125HR is the best performing high resolution FTIR spectrometers commercially available. Compared to the IFS 120M it has (a) a more stable instrumental line shape and (b) a by 30% higher signal to noise ratio. In Schneider and Hase (2007) it is shown that a stable ILS is an important requisite to reach total O₃ precision of better than 1–2%. To guarantee highest quality of our FTIR O₃ products in this work, we only use O₃ amounts inverted from IFS 125HR measurements. The Izaña FTIR also participates in TCCON (Total Carbon Column Observing Network; <http://www.tcon.caltech.edu/>). TCCON is a currently constituting network of FTIR sites with highest requests on quality. It aims on the detection of total amounts of greenhouse gases with a precision of better than 0.1%.

At the Izaña Observatory clean air and clear sky conditions are prevailing around all the year. Firstly, it is located in the region below the descending branch of the Hadley cell, typically above a stable inversion layer. Secondly, it is situated on an island far away from any significant industrial activities. Consequently it offers excellent conditions for atmospheric observations by remote sensing techniques and it is predestinated for calibration and validation activities. Due to its geographic location it is in

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particular valuable for the investigation of stratosphere-troposphere exchange associated with the subtropical jet (e.g. Kowol-Santen et al., 1999; Cuevas et al., 2007) and large scale transport from the tropics to higher latitudes. Figure 1 shows the evolution of the total O₃ amounts in 2005 and 2006 as measured by the site standard Brewer #157, traveling standard Brewer #185, and the FTIR instrument. The Brewer #185 started its operation in April 2005. Further gaps in the time Brewer data are mainly due to intercomparison campaigns of the traveling standard Brewer #185: in September it was in Huelva, Spain; in March/April 2006 and January/February 2007 in Sodankylä, Finland. The gap in December 2005 is due to the (sub-)tropical storm Delta, which hit the island on 28th of November. Peak gusts around 250 km/h were measured on this day and the Brewer #157 suffered some damage and was out of operation during the whole month of December 2005. Other gaps are due to power breakdown after snow storms in winter. The FTIR measures typically three times per week. The gap in February 2005 and the relatively sparse data in winter 2006 are due to snow storms or bad weather conditions.

4 Brewer #157 versus Brewer #185

In this Section we compare total O₃ data from site standard Brewer #157 and the traveling standard Brewer #185. Between January 2005 and February 2007 we find 4300 measurements performed simultaneously by both instruments. The correlation of these O₃ data is shown in Fig. 2. We find a correlation coefficient ρ of 0.998, a regression line slope of 0.996, and an offset of the regression line of 1.5 DU. The scattering around this regression line provides an estimate of the precision of the Brewer instrument with respect to O₃ amounts: The difference between both Brewer instruments is $0.2 \pm 0.4\%$. If we assume that the leading error sources of the O₃ amounts obtained from the two Brewer instruments are independent, we can estimate the precision of one Brewer instrument to $0.4/\sqrt{2}=0.3\%$. However, it is likely that some error sources of two experiments which apply the same technique are correlated. Consequently the precision of 0.3% is only a best case estimation.

4.1 Temporal evolution

In the following we investigate how the difference between both Brewers evolves with time. The left panels of Fig. 3 depict the difference for all individual coincidences versus time. When contemplating this plot it is important to remind the scale on the y-axis. It is given in percent and consequently we are looking on temporal variations in the range of a few permil. Since the noise in the data is also situated in the permil range we can only expect to make some useful observations by contemplating averages over a certain amount of data. Here we average data over 3 months in the sense that we apply data within a radius of 1.5 month around the considered date. The statistics of such averaged data are shown in the right panels. The black bars represent the area where the mean values of the 3-months ensembles are situated with a probability of 95%. The grey bars represent the standard deviation of the 3-month ensembles. These grey bars can be interpreted as overall precision of Brewer #157 and #185 within a three months period. The upper panels show the situation if all 4300 coinciding Brewer measurements are applied. In this case even for the three months averages there is no significant temporal variation of the difference between the two Brewers. It seems that both instruments produce very consistent data even over several years. A small difference between 2005 and 2006 is that the overall precision of Brewer #157 and #185 for 3 months periods is slightly better in 2006 compared to 2005 (0.4% compared to 0.5%; see grey bars).

In addition we perform a separate analysis for low slant column amounts. This is done for two reasons: (1) FTIR measurements are generally performed at relatively low slant column amounts (75% of all FTIR measurements at solar elevation angles above 35°). On the other hand, Brewer measurements are performed during the whole day, and, thus, Brewer data include many O₃ amounts deduced from high slant column amounts. The separate analysis of low slant column assures that the Brewer data are characterised under similar condition as the FTIR data. (2) Systematic ETC errors are amplified for low slant column amounts, since a systematic error in the ETC

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produces a systematic bias in the retrieved slant column amounts (see Eq. 6). The bottom panels of Fig. 3 shows data only if the respective measurement was made for a slant column lower than 400 DU. This slant column amount corresponds to an solar elevation angle of typically above 50°. Consequently, even at the subtropical site of Izaña, these data are not available in winter and the time series is limited to data from February to November. In these graphics we can observe a slight difference between 2005 and 2006. In 2005 both Brewers measure nearly the same, whereas in 2006 the standard size Brewer #157 produces O₃ amounts, which are around 0.5% lower than the Brewer #185 amounts. This observation is robust since it bases on more than 700 coincidences between the Brewers. The areas where the mean difference is situated with a probability of 95% (indicated by black bars in the left panels of Fig. 3) are separated for the years 2005 and 2006: we observe a slight systematic difference in the Brewers' performance between 2005 and 2006.

4.2 Dependence on slant column amounts

In the following we analyse in more detail the differences between the years 2005 and 2006. Therefore, we have a closer look on the dependence of the observed differences on the present slant column amounts. Figure 4 shows this dependence for the two periods: April 2005 to November 2005 and December 2005 to January 2007. The left panels show the individual data points. The right panels show statistics of the averages. Here the averages are made applying all data situated within a radius of 12.5% of the slant column amount, i.e. for a slant column amount of 500 DU we calculated the average of all data between 437.5 and 562.5 DU and for a slant column amount of 1000 DU we apply data between 875 and 1125 DU. The upper panels show the situation for the first period (1100 coincidences). There is a weak dependence of the difference between both Brewers on the slant column amount. For very low slant column amounts the Brewer #157 produces systematically larger amounts than the Brewer #185 (up to 0.5%) and for slant columns above 350 DU the sign changes and the #157 values are smaller than the #185 values. The lower panels show the situation

for the year 2006 (period between December 2005 and January 2007; 3200 coincidences). It differs from the year 2005. Now for low slant column amounts the Brewer #157 data are 0.4% lower than the #185 amounts, and for slant columns above 600 DU both Brewer agree nearly perfectly. Furthermore, we found that in 2006 the data are less noisy than in 2005. These plots show that the inconsistencies between the Brewers with respect to 2005 and 2006 are reflected in a different dependence on the slant column amounts. Such a dependence indicates that there are weak inconsistencies in the ETCs applied for the Brewers (see Eq. 6). The dependence on the slant column amount is in particular large for 2005, indicating larger inconsistencies between both Brewers than in 2006.

5 FTIR (Barret et al., 2002) versus FTIR (Schneider and Hase, 2007)

In this Section we compare total O₃ data from two different FTIR retrieval approaches: a first similar to Barret et al. (2002) (in the following called BA02) and a second similar to Schneider and Hase (2007) (in the following called SH07). By this means we perform an internal consistency check of the FTIR data. Figure 5 shows the correlation between the two approaches. Both retrieval apply the same O₃ absorption signatures but slightly different retrieval strategies. The first consists in an optimal estimation of O₃ profiles alone and the second in a joint optimal estimation of O₃, ⁵⁰O₃/⁴⁸O₃, and temperature profiles. A correlation coefficient of “only” 0.984 is relatively low. The agreement is poorer if compared to the agreement between both Brewer instruments. This is mainly due to errors in the BA02 data. In Schneider and Hase (2007) it was shown that the SH07 approach provides for significantly more precise data than the BA02 approach. It nearly eliminates the error due to uncertainties in the applied temperature profiles, which are the dominant error source of the BA02 approach. On the other hand, the SH07 approach is more sensitive to errors due to ILS uncertainties (Schneider and Hase, 2007). By comparing the data of both approaches we expect to get some information about the stability of the ILS from 2005 to 2007.

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5.1 Temporal evolution

The temporal evolution of the difference of the two FTIR retrieval approaches is depicted in Fig. 6. Like for Fig. 3 the left panels show the individual measurements and the right panels show the statistics for the 3-months averages: the black bars represent the range of the mean values at a 95% confidence level, the grey bars represent the standard deviation. The upper panels show all data and the bottom panels only data for slant column amounts above 400 DU. First of all there is much more noise on the data when compared to Fig. 3. The relatively large noise is mainly due to temperature errors present in the BA02 approach. Again it should be remarked that the BA02 approach does not fully exploit the potential of the FTIR measurements. It is not an optimised approach, but we can exploit its different ILS sensitivity to perform an internal estimation of the FTIR's ILS characterisation. This internal estimation is achieved by averaging over a certain amount of data. This process significantly reduces the temperature error, which is mainly a random error. The remaining systematic signal should then be due to a systematic error source, for which both retrieval approaches have different sensitivities: e.g. the ILS error. The statistics of 3-months averages are depicted in the right panels of Fig. 6. We observe some systematic differences between the years 2005 and 2006. First, the year 2005 is much more variable, which may indicate that the ILS of the FTIR instrument is less stable than in 2006. Second, the difference between both approaches is larger in 2005 than in 2006 (1.5 compared to 0.8%). Assuming that the amplitude of the difference follows the amplitude of the remaining ILS error means that in 2005 the ILS is poorer characterised than in 2006. The bottom right panel of Fig. 6 shows the situation for slant column amounts below 400 DU. Here the differences are especially large in October 2005 (up to 2.2%). Furthermore, we observe that the difference increases from April to October 2005. We think that these observations indicate a gradual decrease of the ILS performance during 2005.

There are several possible explications for the relatively poor instrumental stability in 2005. In January 2005, after the installation of the instrument, the ILS was very

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well characterised (difference between BA02 and SM07 data of 0%). However, in the following there was a sequence of experimental complications. The setting of the fundam
ent or the whole surface below the container may have still been in progress during the
first months of the measurements. In February 2005 there was a snow storm in
Izaña and a subsequent breakdown of the power supply for several days. As a conse
quence the temperature in the container fell down to around 0 C°. These mechanical
and thermal stresses may have produced a degradation of the optical alignments. Fur
thermore, the low temperature damaged the hygroscopic entrance windows which had
to be replaced. Unfortunately we only performed two independent ILS calibration mea
surements between January and November 2005. This is definitively not sufficient for a
high quality characterisation of the ILS, in particular if we consider the aforementioned
adverse conditions. In November 2005 we installed a new firmware and reinstalled
the detectors for technical servicing. The latter significantly reduced a channeling of
0.58 cm⁻¹. Since December 2005 the instrument is operating continuously and there
were no further modification necessary. In particular there was no further significant
temperature breakdown inside the measurement container. Since the end of 2005 it is
kept continuously at 22 C°.

5.2 Dependance on slant column amounts

The ILS error of the FTIR O₃ data slightly depends on the O₃ slant column amount
(Schneider and Hase, 2007). The error is larger for low slant column amounts than
for high slant column amounts. Since the BA02 and SH07 approach have different ILS
error sensitivities, this dependance should be observable in the difference of BA02 and
SH07 O₃ amounts. Figure 7 depicts the dependance of the difference between BA02
and SH07 O₃ on the slant column amount. In 2005 (upper panels) we observe a clear
increase of the negative difference for low slant column amounts, whereas in 2006 this
increase is much smaller. Thus, Fig. 7 confirms our conclusions drawn from Fig. 6: in
2005 the FTIR instrument is poorer characterised than in 2006.

6 Brewer versus FTIR

In this Section we compare total O₃ measurements of Brewer and FTIR. To exclude influences due to temporal variabilities we require that both Brewer and FTIR measurements should coincide within 30 min. Between January 2005 and February 2007 we identified a total of 305 FTIR measurements which fulfill these coincidence criteria with the Brewer measurements: 240 for the Brewer #157, 165 for the Brewer #185. Both Brewer instruments measure generally during the whole day. Figure 8 depicts the correlation between the site standard Brewer #157 and the FTIR O₃ amounts. The left panel shows the correlation between the Brewer data and the FTIR data obtained from the BA02 approach. The agreement between the Brewer and these FTIR data is quite good: a correlation coefficient of 0.982 and a difference of $4.0 \pm 1.1\%$. This agreement is much better than reported by other studies (e.g. Barret et al., 2002; Schneider et al., 2005). It demonstrates the high quality of the Brewer and FTIR measurements performed at Izaña. In particular, it confirms the potential of the FTIR technique when the instrumental aspects of the recipe as presented in Schneider and Hase (2007) are considered.

However, there is still a large potential to further improve the agreement. As shown in Schneider and Hase (2007) the BA02 error budget is dominated by errors in the assumed temperature profiles and applying the SM07 approach should widely reduce the overall FTIR errors. The correlation of the FTIR SH07 data with the Brewer #157 data is shown in the right panel of Fig. 8. It yields a correlation coefficient of 0.992. The difference between both instruments is $4.9 \pm 0.7\%$. The SM07 data agree significantly better with the Brewer data than the BA02 data. However, according to Figs. 3 and 6 we find differences in the performance of the Brewer and FTIR instruments for the years 2005 and 2006. There are inconsistencies in the data of the different years. Furthermore, both the Brewer and, in particular, the FTIR instrument perform better in 2006 than in 2005. The reasons are discussed in Sect. 4 and 5. Consequently the year 2006 is much more representative for the real potential of the Brewer and FTIR

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instruments. If we only correlate Brewer and FTIR SH07 data measured after November 2005 we get a correlation coefficient of 0.996 and a difference of $-4.7 \pm 0.5\%$. The BA02 approach, on the other hand, only yields a correlation coefficient of 0.984 and a difference of $-4.0 \pm 1.0\%$: for the BA02 approach the agreement does not improve significantly if we limit the comparison to data measured after November 2005. The BA02 data are dominated by errors caused by the assumed temperature profiles, which are much larger than the errors caused by inconsistencies in the instrument's performance. The SH07 approach produces a standard deviation of the difference Brewer-FTIR of 0.5%. This is an excellent value for two independent remote sensing experiments performed over more than 1 year. It can be interpreted as the root-square-sum of the precisions the Brewer and FTIR instrument. As aforementioned the Brewer precision is not better than 0.3%. Consequently the Brewer/FTIR comparison is the empirical proof that FTIR measurements can provide O_3 amounts with a precision of better than 0.4% over several years. Within all 240 coincidences we found one day which clearly lies out (2 March 2006; marked by blue circle in both panels of Fig. 8). So far we have no explication for this outlier. We do not consider this data point for the following analysis since it is not representative but would widely influence the results.

The Brewer-FTIR comparison confirms the theoretical prediction of [Schneider and Hase \(2007\)](#). However, there are significant systematic differences between both datasets: The FTIR observing system measures systematically 4.7% more O_3 than the Brewer system. Systematic differences may be produced by errors in the applied spectroscopic parameterisation. The HITRAN data ([Rothman et al., 2005](#)) applied by the FTIR algorithm are assumed to have an accuracy of around 2%. If we make a similar systematic error assumption for the [Bass and Paur \(1985\)](#) cross sections applied by the Brewer algorithm our observation would exceed the range expected from inconsistencies in the applied spectroscopic data. Consequently, these error assumptions are either too optimistic or the observed differences are partly due to other systematic errors sources, like errors in the characterisation of the instruments.

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6.1 Temporal evolution

Figure 9 shows the temporal evolution of differences between Brewer #157 and the FTIR data obtained by the SM07 approach. The upper panels show the data from all 240 coinciding measurements. Here the 95% confidence ranges (black bars in right panels) are much broader than in Fig. 3 due to a much smaller number of individual measurements applied for averaging. We observe clear differences between 2005 and 2006. In March 2005 FTIR O₃ amounts are typically 5.7% higher than Brewer #157 amounts, while in 2006 these differences are typically 4.6%. We observe a continuous increase of the Brewer #157 O₃ with respect to the FTIR O₃ between March 2005 to April 2006. The stability of the agreement since the end of 2005 is remarkable. Both techniques agree within 0.5% during more than 14 months. This excellent agreement is also achieved in 2005 if we consider only 3-month periods (see grey bars in right panels). Only in January and October/November 2005 it is larger. In both situations the ensembles include data for different FTIR and Brewer instrumental characteristics: the internal FTIR check (Fig. 6) shows steps in February and November 2005 and the internal Brewer check (Fig. 3) shows a step in December 2005. The same excellent agreement can be observed if we limit to slant column amounts below 400 DU (bottom panels). Within 3-month periods the data agree always within 0.5%. Concerning the differences between 2005 and 2006 the continuous increase during 2005 is even more pronounced if compared to the upper panels (from -6.0% to -4.6%). Finally, Fig. 10 depicts the temporal evolution of the difference between Brewer #185 and FTIR. As before, we observe a clear difference between 2005 and 2006. In 2005 the FTIR measures typically around 5.5% larger amounts than the Brewer #185. In 2006 the difference reduces to typically 4.8%.

Figures 9 and 10 have many similarities but also two differences. These are: (a) the #185-FTIR data are more noisy than the #157-FTIR data, i.e. the #185 data are more noisy than the #157 data. This was already observed during the ETC calibration in October 2005 and is due to an inappropriate filter applied by the Brewer #185 for high

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solar fluxes (K. Lamb, private communication). (b) The difference between 2005 and 2006 is slightly smaller for the #185-FTIR data compared to the #157-FTIR. A reason could be that the differences between 2005 and 2006 as observed in #185-FTIR are mainly due to inconsistencies in the FTIR data and the differences observed in #157-FTIR are due to both inconsistencies in the FTIR and the Brewer #157 data.

6.2 Dependence on slant column amounts

Figure 11 shows the dependence of the difference of the FTIR and Brewer #157 on the slant column amounts. The upper panels show the situation for all 106 coincidences between January 2005 and November 2005. For increasing slant column amounts we observe a clear increase of the Brewer #157 O₃ amounts with respect to the FTIR amounts. At low slant columns the Brewer measures amounts which are 5.6% lower than the FTIR amounts. For slant column amounts at 700 DU this difference reduces to 4.4%. For the 2006 period the dependence on the slant column amount is less pronounced: at 700 DU the difference is around 4.5% and below 450 around 4.8%. This means that the FTIR and Brewer #157 data are more consistent in 2006 than in 2005. The upper right panel of Fig. 11 has significant similarities with the right upper panel of Fig. 7. This suggests that the inconsistencies between the FTIR and Brewer #157 are mainly due to errors in the FTIR data.

Figure 12 shows the same as Fig. 11 but for the difference between FTIR and Brewer #185. We also found that dependence on slant column amounts is larger in 2005 if compared to 2006.

7 Some remarks on the excellent quality of Izaña's O₃ measurements

To our knowledge there is world-wide no other observatory measuring total O₃ by ground-based techniques at the high precision as presented in this work. The reasons for this are manifold: (a) the outstanding meteorological conditions that prevail at

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the Izaña Observatory. For example, the high quality of the FTIR data is only achieved since there are no or only negligible intensity fluctuations over short time scales. The typical scan time for one FTIR measurement is in the order of 10 min. Intensity fluctuation during this time produce artificial intensity offsets in the spectra and ILS distortions.

Intensity fluctuations are more frequent at less favorable measurement sites due to changes of the atmospheric transparency (due to clouds, short-scale inhomogeneities in water vapour, tropospheric aerosols, contrails, ...). They are one of the major remaining error sources. (b) The application of state-of-the-art instrumentation. The Brewer triad is formed by three state-of-the-art instruments with double monochromator. The FTIR system consist of a very precise solar tracker [Huster \(1998\)](#), a Bruker IFS 125HR (stable ILS), and applies photo-voltaic detectors. These are the most important instrumental requirements needed for high precision FTIR measurements. Instabilities in the ILS are one of the dominant remaining error sources. We think that these instabilities are responsible for the internal FTIR inconsistencies observed during 2005. (c) The high precision of both Brewer and FTIR depends on regularly performed calibration measurements. In case of the Brewer the ETC and wavelength are monitored internally by calibration lamps several times per day. Once per year the ETC is transferred from the traveling world standard Brewer. The slit function remains very stable. It is measured every year. In case of the FTIR the ILS calibration by low pressure gas cells ([Hase et al., 1999](#)) is since November 2005 performed every 2–3 months. (d) Finally, it is important to apply an optimised retrieval algorithm: the FTIR only reaches a precision better than 0.5% if the retrieval strategy as proposed by [Schneider and Hase \(2007\)](#) is applied.

8 Summary and conclusions

At the Izaña Observatory the two most precise ground-based total O₃ monitoring instruments (Brewer spectrometer with double monochromator and high resolution FTIR spectrometer) are continuously measuring side-by-side. Our study shows that the

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precision of Brewer and FTIR are quite similar. The agreement between both techniques is much better than reported in previous studies. Our study confirms empirically, that both Brewer and FTIR can achieve a precision of around 0.4%.

The high precision provides a unique opportunity of a detailed investigation of the performance of both techniques. In general we found that Brewer and FTIR are very stable instruments, even over extended periods and, concerning the Brewer, even after transportation to and from different campaign sites. A detailed analysis of averaged data allows us to reveal slight inconsistencies between the 2005 and 2006 data. In case of the Brewer we think that the ETC determined for the Brewer #157 for the 2005 period is slightly less accurate than for the 2006 period. However these errors are very small and are in agreement with the expected ETC uncertainties. Our study demonstrates a very high precision of Brewer spectrometers with double monochromators, much better than what can be achieved by instruments with single monochromator (Fioletov et al., 2005). State-of-the-art Brewer instruments combine high precision with mobility. They are perfectly suited for calibrating O₃ instruments at different sites. Furthermore, Brewer spectrometers are relatively economic and measure O₃ throughout the day and during the whole year. Concerning the FTIR data we found that in the year 2005 the ILS is relatively poorly characterised. The period from January to November 2005 is not representative for the real potential of the FTIR technique and the 2005 FTIR data does not reach the quality as predicted in Schneider and Hase (2007). The reasons for this are listed in Sect. 5. But even then we found an agreement to the Brewer within 0.8%. Since December 2005 the FTIR instrument is very well characterised. For this period it agrees with the Brewer within 0.5%, which confirms the prediction of Schneider and Hase (2007). This supports the theoretical quality assessments of other FTIR products, for which no direct empirical verification can be achieved. This result is of particular interest in the context of ground-based high precision measurements of greenhouse gases, which are performed within TCCON (Washenfelder et al., 2006) and will serve for the validation of OCO (Crisp et al., 2004). TCCON aims on a precision of 0.1%.

We find a significant systematic difference between the FTIR and Brewer data of

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4–5%. This difference could be caused by systematic errors in the FTIR data due to a combination of systematic ILS uncertainties and systematic errors in the HITRAN parameterisation. However, the Bass and Paur (1985) cross sections applied by the Brewer algorithm could also be responsible. This needs further investigation and should be addressed in a future work.

A currently very important issue is the significant difference of up to 10%, that is observed at polar regions between ground- and satellite-based measurements. These differences are still not understood (Bojkov et al., 2006) and a detailed investigation of the precision and accuracy of ground-based O₃ data measured for large slant column amounts is very important. Izaña's instrumentation can make valuable contributions to investigate this issue. In this context, we plan a special campaign where we will perform FTIR measurements during several days from the sunrise to the sunset. We hope that this campaign will give a better insight in the differences between the FTIR and Brewer data and its dependence on slant column amounts.

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Table 1. Main differences between Brewer and FTIR experiments.

	Brewer	FTIR
number of channels/spectral bins	4	7500
resolution power	6×10^2	2×10^5
measured wavelength regions	UV	mid-infrared
line-of-sight	“simplified” considerations	ray tracing model
field-of-view	2.7° (larger than solar disc)	0.2° (smaller than solar disc)
wavelength calibration	important: due to low resolution of measured spectra and very fine-structured σ_{O_3}	not necessary: high resolution spectra is self-calibrating
slit function (or ILS) calibration	important to calculate the convolved σ_x	important to correctly simulate convolved spectra
intensity calibration	not important: algorithm bases on ratio of channels	not necessary: highly resolved measurements are self-calibrating with respect to spectra broadband structures not required
determination of ETC	is determined from a set of measurements (Langley plot or transferred from standard instrument)	
T dependance of σ_{O_3}	not considered (uses effective σ_{O_3})	fully considered as well as pressure dependency of σ_{O_3}
measurement frequency	very high, every 15 min during whole day (for $\Theta < 83^\circ$)	only 1–2 measurements per day, on typically 3 days per week
O ₃ isotopologues	no separation of distinct isotopologues	measures $^{48}\text{O}_3$, asymmetric and symmetric $^{50}\text{O}_3$ and $^{49}\text{O}_3$ individually
size/mobility	small instrument, highly-mobile, best-suited as mobile reference instrument	large instrument, installed inside container, limited mobility
price	150 kEUR	500 kEUR

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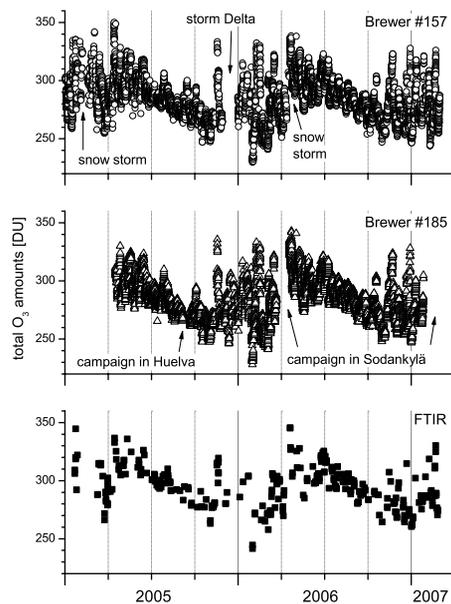


Fig. 1. Time series of total O₃ as measured by the Brewer site standard Brewer #157, the traveling standard Brewer #185, and the FTIR instrument.

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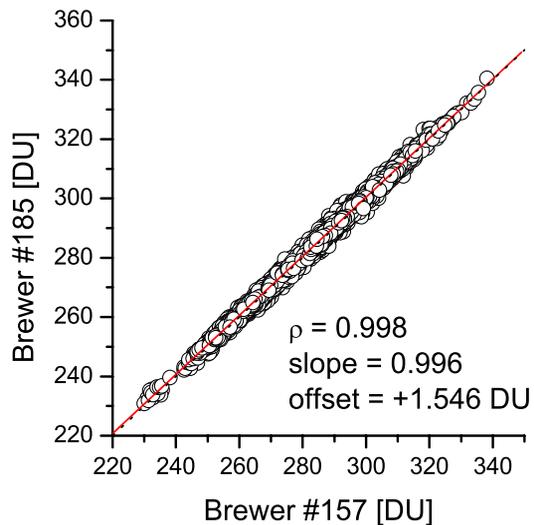


Fig. 2. Correlation between total column amounts of Brewer #157 and Brewer #185. Black circles are individual measurements, red line represents linear regression line of least squares fit.

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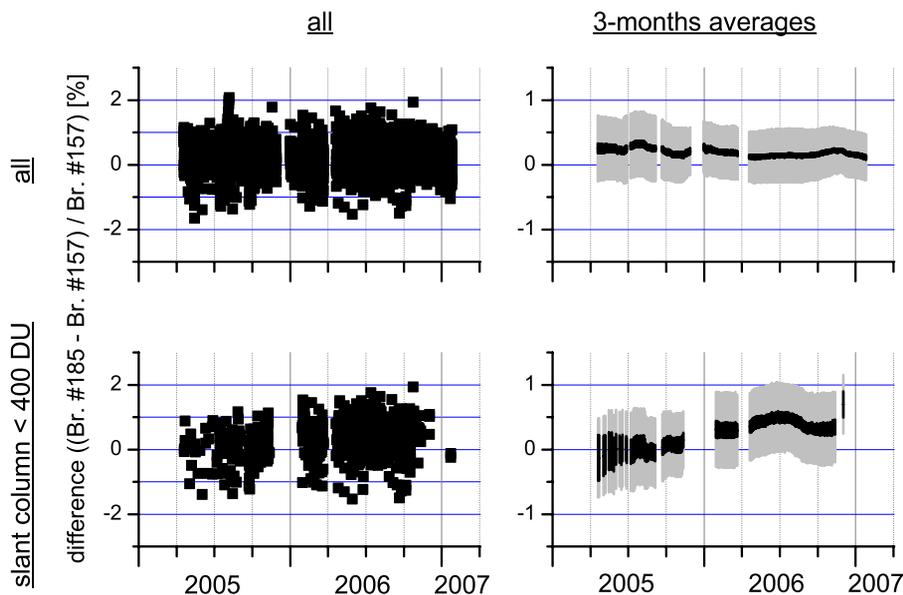


Fig. 3. Time series for the differences of total O_3 measured by the Brewer #157 and the Brewer #185 ($(\#185 - \#157) / \#157$). Upper panels: for all coinciding measurements; bottom panels: for measurements with low slant columns (<400 DU). The left panels show all individual coincidences and the right panels the statistics of three months averages: the black shaded area covers the range within which the mean value is situated with a probability of 95%; the grey-shaded area indicates the standard deviation. The scale of the y-axis of the right panels is extended by a factor 2.

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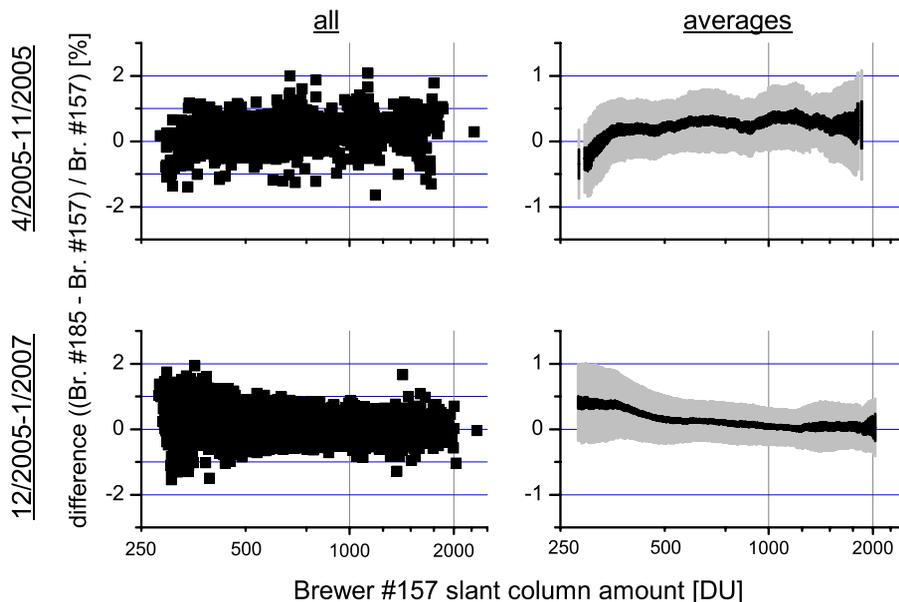


Fig. 4. Differences of total O_3 measured by the Brewer #157 and the Brewer #185 ($(\#185 - \#157) / \#157$) versus slant column amount. Upper panel: for measurements between April and November 2005; bottom panel: for measurements between December 2005 and January 2007. The left panels show all individual coincidences and the right averages over values within a radius of 12.5% of the slant column amount: the black shaded area covers the range within which the mean value is situated with a probability of 95 %; the grey-shaded area indicates the standard deviation. The scale of the y-axis of the right panels is extended by a factor 2.

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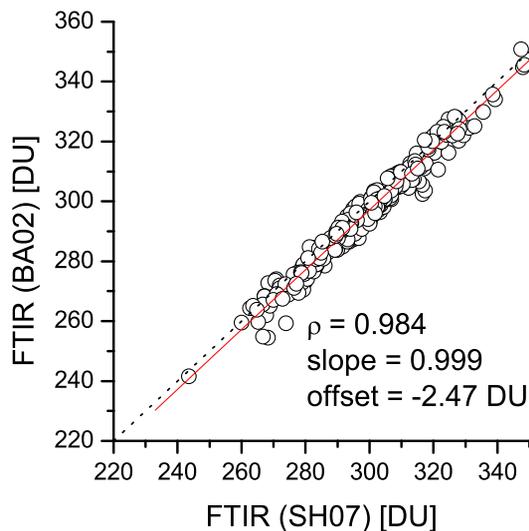


Fig. 5. Correlation between total column amounts for different FTIR approaches (a first similar to Barret et al. (2002) and a second according to Schneider and Hase (2007)). Black circles are individual measurements, red line linear regression lines of least squares fit.

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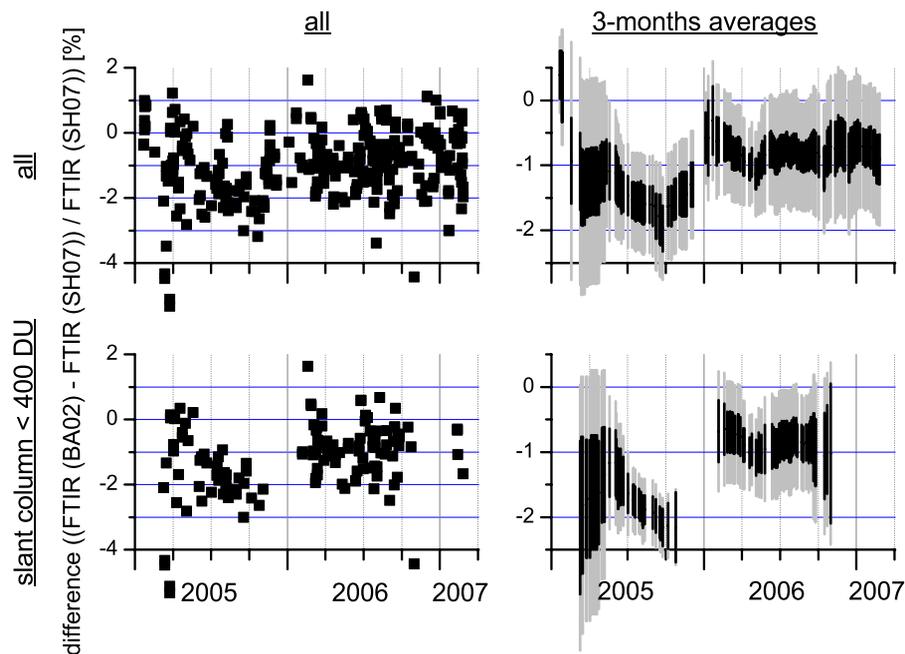


Fig. 6. Time series for the differences between the different FTIR approaches (FTIR (BA02)–FTIR (SH07)) / FTIR (SH07)). Content of panels, symbols, and colours is the same as in Fig. 3.

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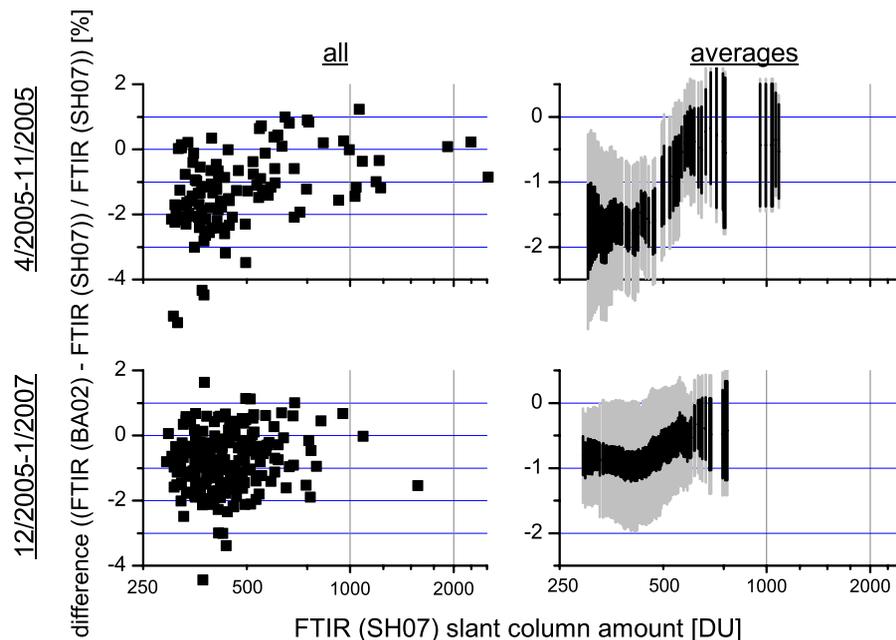


Fig. 7. Differences of total O_3 measured by the different FTIR approaches (FTIR (BA02)–FTIR (SH07)) / FTIR (SH07)) versus slant column amounts. Content of panels, symbols, and colours is the same as in Fig. 4.

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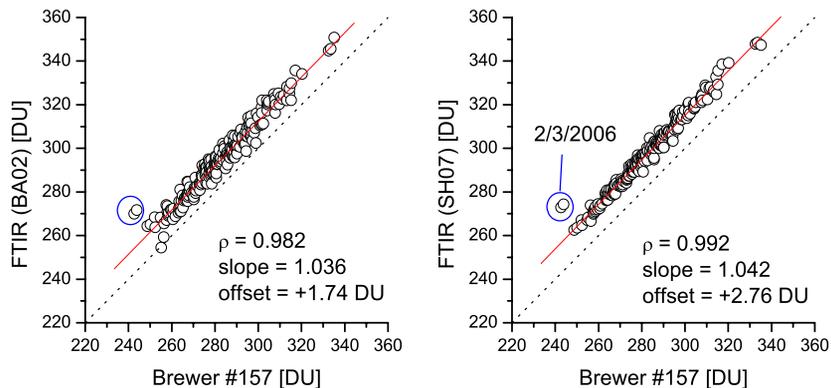


Fig. 8. Correlation between total column amounts of Brewer #157 and FTIR. Black circles are individual measurements, red lines linear regression lines of least squares fits. Left panel: correlation between Brewer and BA02 FTIR retrieval; right panel: correlation between Brewer and SH07 FTIR retrieval.

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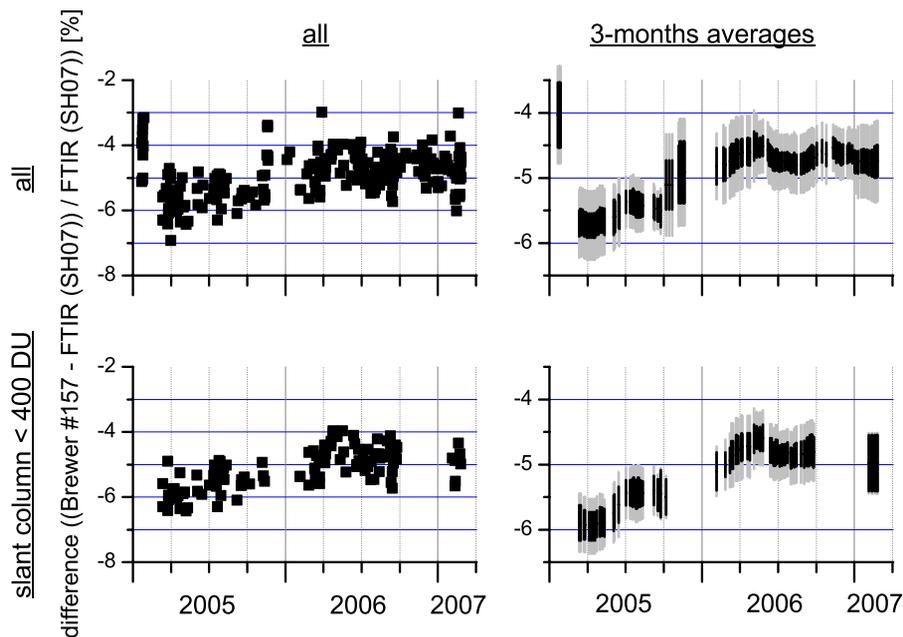


Fig. 9. Time series for the differences of total O_3 measured by the Brewer #157 and the FTIR. $((\#157 - FTIR) / FTIR)$. Content of panels, symbols, and colours is the same as in Fig. 3.

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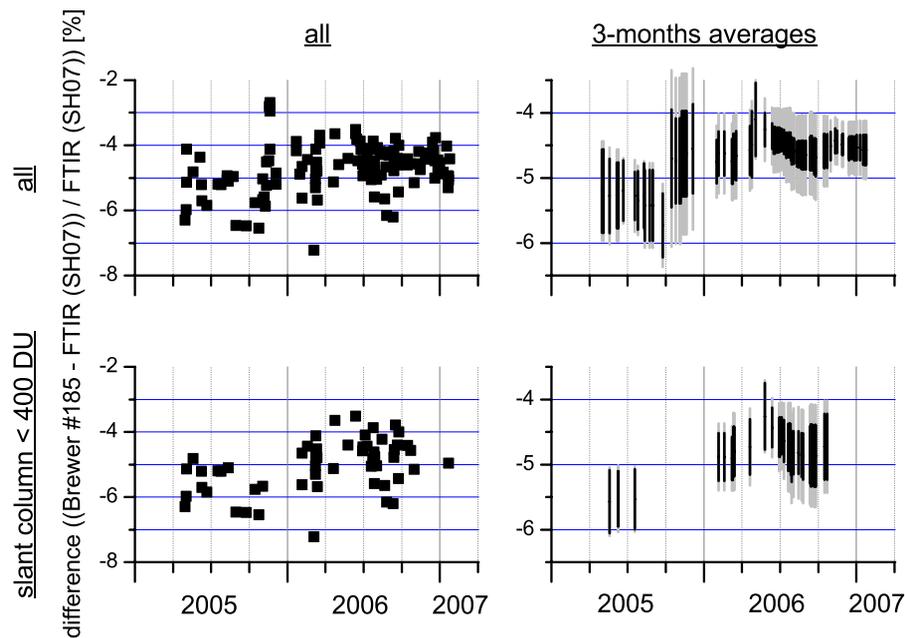


Fig. 10. Time series for the differences of total O_3 measured by the Brewer #185 and the FTIR. ($\#185 - FTIR$) / $FTIR$). Content of panels, symbols, and colours is the same as in Fig. 3.

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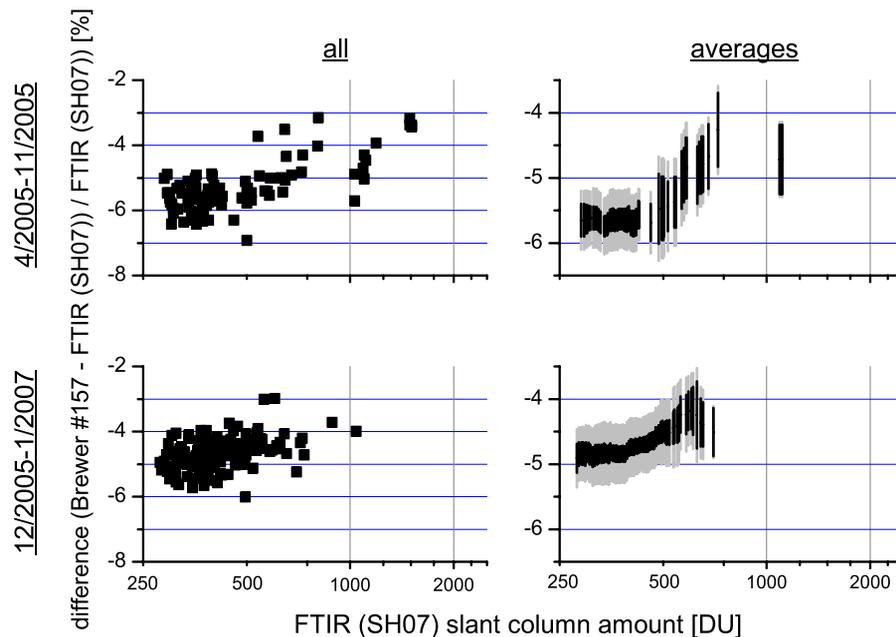


Fig. 11. Differences of total O_3 measured by the Brewer #157 and the FTIR. $((\#157 - \text{FTIR}) / \text{FTIR})$ versus slant column amounts. Content of panels, symbols, and colours is the same as in Fig. 4.

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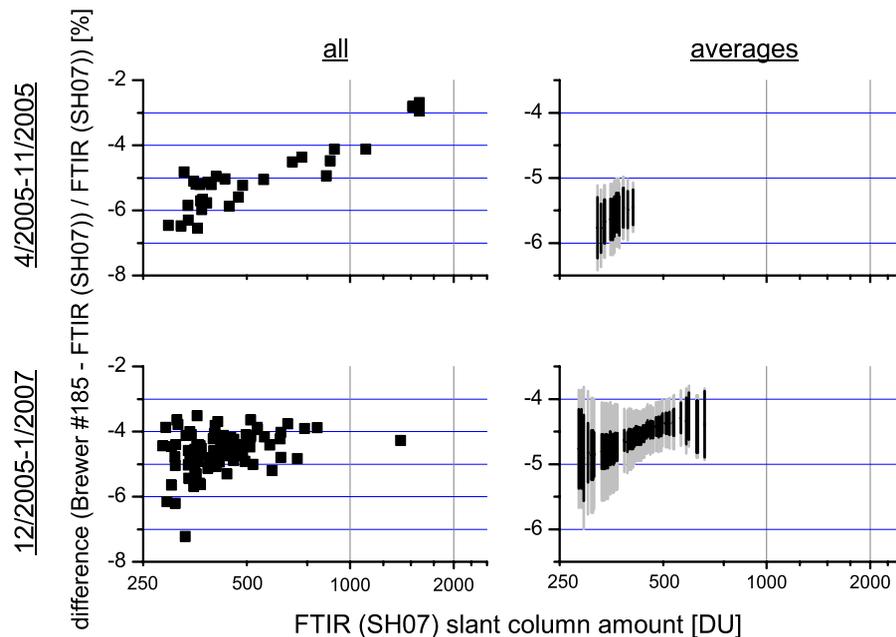


Fig. 12. Differences of total O_3 measured by the Brewer #185 and the FTIR. $((\#185 - \text{FTIR}) / \text{FTIR})$ versus slant column amounts. Content of panels, symbols, and colours is the same as in Fig. 4.

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