Stability of the Regional Brewer Calibration Center for Europe Triad during the period 2005 – 2016

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Abstract. Total ozone column measurements can be made using Brewer sprectrophotometers which are calibrated periodically in intercomparison campaigns with respect to a reference instrument. In 2003, the Regional Brewer Calibration Centre for Europe (RBCC-E) was established at the Izaña Atmospheric Research Centre (Canary Islands, Spain) and since 2011 it transfers its own calibration, mainly to other European Brewers, using the Brewer #185 as travelling reference. This work is focused on reporting on the stability of the measurements of the RBCC-E Triad (Brewers #157, #183 and #185) made at the Izaña Atmospheric Observatory during the period 2005 – 2016. In order to study the long-term precision of the RBCC-E Triad, it must be taken into account that each Brewer performs a large number of measurements every day and, hence, it becomes necessary to calculate a representative value of all of them. This value was calculated from two different methods previously used to study the long-term behaviour of the World Reference Triad (so-called Toronto Triad) and Arosa Triad. Applying their procedures to the data from the RBCC-E Triad allows the comparison of the three instruments. In daily averages, applying the procedure used for the World Triad Reference, the RBCC-E Triad presents a relative standard deviation equal to σ=0.41% which is calculated as the mean of the individual values for each Brewer ($\sigma_{157} = 0.362\%$, $\sigma_{183} = 0.453\%$ and $\sigma_{185} = 0.428\%$). Alternatively, using the procedure used to analyze the Arosa Triad, the RBCC-E presents a relative standard deviation of about $\sigma = 0.5\%$. In monthly averages, the method used for the data from the World Triad Reference give a relative standard deviation mean equal to σ=0.3% ($\sigma_{157} = 0.33\%$, $\sigma_{183} = 0.34\%$ and $\sigma_{185} = 0.23\%$). Whereas, the procedure of the Arosa Triad gives monthly values σ=0.5%. In this work, two ozone datasets are analyzed: the first include all the ozone measurements available while the second only includes the simultaneous measurements of all three instruments. Furthermore, this paper also describes the Langley method used to determine the Extra-Terrestrial Constant (ETC) for the RBCC-E Triad, the necessary first step toward accurate ozone measurement. Finally, the short-term, or intraday, stability is also studied to identify the effect of the solar zenith angle on the accuracy of the RBCC-E Triad.

1 Introduction

The ozone layer is a region of the Earth's stratosphere that absorbs most of the Sun's Ultraviolet (UV) radiation (Anwar et al., 2016). Historical measurements -pre 1980- indicated that the morphology of ozone was not changing significantly with time.

However, the Antarctic measurements of Farman et al. (1985) changed that view. Concerns related to the negative effects that UV can have on terrestrial life led to the signing of the Montreal Protocol in 1987, where 197 countries agreed to reduce the agents that led to this decrease (Sarma and Andersen, 2011). From this date, the monitoring and control of the total ozone column abundance has been a priority of the World Meteorological Organization (WMO). This task requires instruments that can measure the total ozone column concentration with an accuracy of $\sim$ 1-3% such as the Dobson and Brewer which are considered as ideal instruments for monitoring the ozone abundance (Basher, 1985; Varotsos and Cracknell, 1994; Fioletov et al., 2005; Scarnato et al., 2009).

Brewer ozone spectrophotometers are used to measure the total ozone column (TOC), ultraviolet irradiance (Fioletov, 2002) and, more recently, the aerosol optical depth in the ultraviolet range (Carvalho and Henriques, 2000; Gröbner et al., 2001; López-Solano et al., 2017). This instrument is mounted on an azimuth tracker that determines the TOC from a direct measurement of the solar radiation. The grating system separates the solar radiation and a slit mask mechanism is used to select the different UV bands to be measured which are associated with maximum and minimum ozone absorption cross-sections.

After the development of the first Brewer in the early 1970s (Brewer, 1973; Kerr et al., 1985), it has had on-going technical improvements to improve its accuracy. For example, in the photomultiplier, diffraction gratings, operating software used as well as the incorporation of new measurement routines (Fioletov et al., 2005, 2011; Karppinen et al., 2015; Fountoulakis et al., 2017). However, possibly the greatest improvement has been the transition from a single to a double monochromator. This eliminates the presence of stray light in the measurements which causes a decrease in the TOC at large solar zenith angles (Karppinen et al., 2015). In practice, the single Brewer presents this problem for large ozone slant column (OSC). Although, depending on the instrument, this effect may be greater or lesser (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2015; Redondas and Rodriguez-Franco, 2016).

The calibration of the Brewer is traceable from the World Triad Reference, managed by Environment and Climate Change Canada, consisting of Brewers #008, #014 and #015, and located at Toronto. These single Brewers are calibrated every few years at the Mauna Loa Observatory (Hawaii) using the Langley-technique (Fioletov et al., 2005). A second triad formed by double Brewers #145, #187 and #191 is also operated in parallel with the World Triad Reference (Netcheva, 2014; Fioletov and Netcheva, 2014; Zhao et al., 2016). Also, the Swiss Federal Office of Meteorology and Climatology (Meteo Swiss) has the Arosa Triad formed by the singles (#040 and #072) and double (#156) Brewers (Stübi et al., 2017). However, the Brewers distributed around the World are calibrated by comparison with the travelling standard reference, Brewer #017, managed by International Ozone Services (IOS) and Brewer #158 manufacturer by Kipp & Zonen.

In addition, since November 2003 and within the World Meteorological Organization (WMO) and the Global Atmosphere Watch (GAW) Programme, the Regional Brewer Calibration Centre (RBCC-E) for RA-VI Region was established at the Izaña Atmospheric Observatory (IZO) which is located on the island of Tenerife, managed by the State Meteorological Agency of Spain (AEMET). The RBCC-E is the European Brewer Reference and hence can calibrate and transfer its own calibration. Its trajectory started in the year 1997 when the first double Brewer #157 was installed at IZO, running in parallel with a single Brewer #033 for six months. In January 2005, a second double Brewer, the #183, was installed and designated as the travelling reference. The single Brewer #033 was moved to Santa Cruz Meteorological Station (CMT) in December 1997, leaving the

RBCC-E with only two instruments. In July 2005, a third double Brewer #185 was installed. Since that moment, the RBCC-E has been formed by the Brewers #157, #183 and #185. The TOC measured by Regional Primary Reference #157 are sent regularly to different world data servers. The Regional Secondary Reference #183 is used to developing and test. Whereas, the Regional Travelling Reference corresponds to Brewer #185.

The Izaña Atmospheric Observatory is located on the island of Tenerife, on the top of a mountain plateau at 2373 m a.s.l. The observatory is thus located on the region below the descending branch of the Hadley cell, typically above a stable inversion layer, and on an island far away from any significant industrial activities. This ensures clean air and clear sky conditions around all the year and offers excellent conditions to perform the Langley-technique, except for some days where the Saharan dust intrusions inhibit the measurements of the direct solar radiation. Each Brewer can be calibrated "in situ" and independently using the Langley-technique, without the need to move them to other locations. Moreover, the traceability between the RBCC-E Triad and the World Triad Reference is checked during the calibration campaigns through the travelling references #185 and #017. In this comparison, both instruments agree within 0.5%. These values have been calculated using measurements in a range where Brewer #017 measurements are not strongly affected by stray light (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2015; Redondas and Rodriguez-Franco, 2016)

The RBCC-E also organizes inter-comparisons which are held annually, alternating between Arosa (Switzerland) and El Arenosillo (Spain). Since 2011, more than 150 calibrations have been conduted (Cuevas et al., 2015). In these campaigns, the RBCC-E facilitates a new calibration for each instrument. Moreover, in order to obtain an ozone value with better accuracy, the RBCC-E advises on the need to reprocess the observations performed by each brewer at its local station (Redondas et al., 2018). Aside from regular inter-comparisons, the RBCC-E has carried out other research campaigns supported by the ESA CalVal project. The NORDIC campaigns, with the objective to study the ozone measurements at high latitudes, and the Absolute Calibration Campaigns performed at IZO with the participation of Brewer and Dobson reference instruments. The participating Brewers and the travelling reference #185 operate with the same schedule throughout these campaigns. The TOC recorded by the travelling reference #185 are used to calibrate the participating Brewers and also to conduct research works (De La Casinière et al., 2005; Redondas et al., 2015; Redondas and Rodriguez-Franco, 2016).

Finally, it should be also mentioned that within the framework of COST Action ES1207, "A European Brewer Network" (EUBREWNET), the RBCC-E and AEMET are developing a dataserver for EUBREWNET (http://rbcce.aemet.es/eubrewnet) which will allow the calculation of the TOC in near real time (Rimmer et al., 2017). This completes the objectives of this COST action, whose aim is establishing a coherent network of Brewer monitoring stations in order to harmonise operations and develop approaches, practices and protocols to achieve consistency in quality control, quality assurance and coordinated operations. Currently, approximately 40 Brewers, mainly European, send their data automatically every 20 minutes to EUBREWNET's dataserver. This dataserver also allows the reprocessing of data for homogenization and automatic quality control.

The present work focused on investigating how similar are the measurements made by the Brewers #157, #183 and #185 each day and how stable does this behaviour remain over time. This allows us to identify periods with lower or higher agreement between the Brewers. The RBCC-E measurements are evaluated from the methods described for the World Reference and

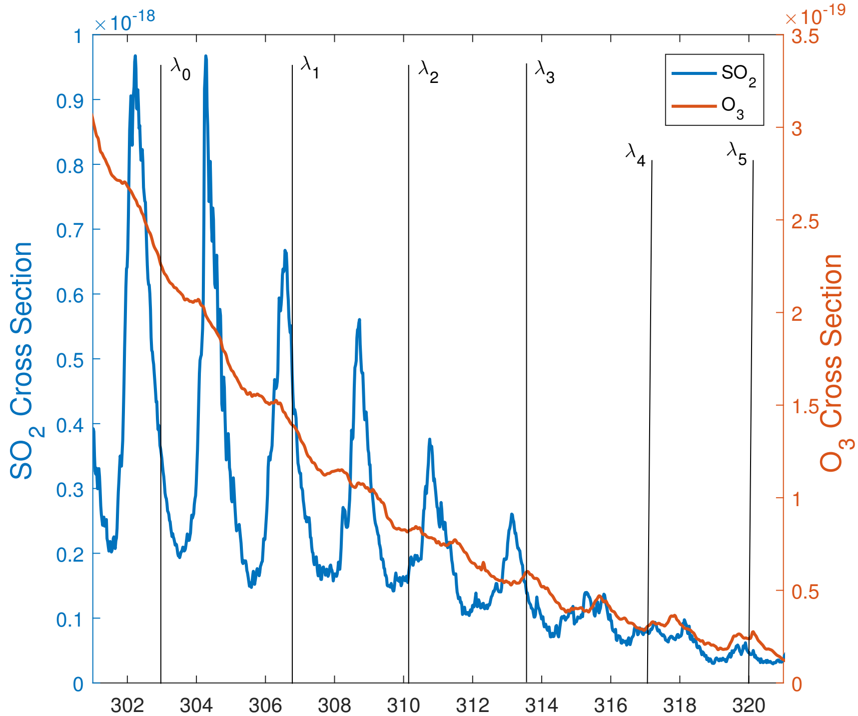

Figure 1. Ozone and sulfur dioxide absorption cross sections. The solar radiation is measured for the intensity bands ($\lambda_{1-5} = 306.4, 310.1, 313.5, 316.8, 320.0 nm$). In contrast, the wavelength $\lambda_0 = 303nm$ is used for a check routine.

Arosa Triads to study its stability. With this idea in mind, this work has been structured as follows: an approach to ozone retrieval and Langley method is presented in Section 2. The ozone values recorded in the period 2005-2016 and datasets used are shown in Section 3. The methods used to calculate the daily ozone value and the results obtained from these values and its discussion are presented in Section 4. Also, this section incluides results of a study on the behaviour of the RBCC-E Triad as a function of SZA range at which the measurements were performed. Finally, the conclusions are presented in Section 5.

2 Theoretical Approach

2.1 Ozone retrieval.

The standard (so-called DS) routine used to determine the TOC from direct sunlight radiation, measuring the signal intensity in five bands ($\lambda_{1-5} = 306.4, 310.1, 313.5, 316.8, 320.0 nm$) which are associated with maximum and minimum O_3 and SO_2 absorption cross sections, see Fig. 1. Despite that SO_2 presents a more efficient absorption, its lower presence in the atmosphere (5 D.U.) compared to the ozone (200-500 D.U.) causes that the greater absorption of UV radiation is due to the latter (Kerr, 2010). The intensity measured F,in raw counts for each wavelength, can be expressed in terms of counts per second, after applying some instrumental corrections (dark counts, dead time and temperature coefficients) and, also, taking into account the contribution of Rayleigh scattering.

Using standard Brewer operational variables, the TOC can be obtained as follows,

$$O_3 = \frac{MS9 - ETC}{\alpha \cdot \mu} \tag{1}$$

where MS9 (so-called double ratio) is calculated as follows, (Brewer, 1973; Kerr et al., 1981, 1985; Kipp & Zonen, 2008).

$$MS9 = 10^4 \sum_i w_i \log F_i = 10^4 (\log F_2 - 0.5 \log F_3 - 2.2 \log F_4 + 1.7 \log F_5) \tag{2}$$

The ozone absorption coefficient, α, is calculated from dispersion test (Redondas et al., 2014a)

$$\alpha = 10^4 \sum_i w_i \log \alpha_i = 10^4 (\log \alpha_2 - 0.5 \log \alpha_3 - 2.2 \log \alpha_4 + 1.7 \log \alpha_5) \tag{3}$$

where α_i represents the band intensity calculated for each wavelength indicated in Fig. 1.

The weights, $w_i = (-1, 0.5, 2.2, -1.7)$ and the wavelengths, $\lambda_i = (310.1, 313.5, 316.8, 320.0nm)$, used have been especially selected to suppress the aerosol and SO_2 effects in the measured signal (Dobson, 1957; Kerr et al., 1981). These w_i and λ fulfill the equations 4 and 5 ensuring that any linear effects with wavelength are suppressed and also allow to minimize any small shift in wavelength and the influence of SO_2 on the ozone retrieval.

$$\sum_{i=1} w_i = 0 \tag{4}$$

$$\sum_{i=1} w_i \lambda_i \approx 0 \tag{5}$$

It is important to note that the factor 10^4 introduced in Eq. 2 is because the Brewer algorithm works in an internal base 10 logarithmic space multiplied by this factor. A more extensive description about dispersion test and the mathematical procedure to calculate the ozone concentration can be found in (Gröbner et al., 1998; Kipp & Zonen, 2008). Finally, the ETC (so-called Extra-Terrestrial Constant) must be calculated directly using the Langley-technique or transfers by comparison with a reference instrument.

2.2 Langley calibration method for the RBCC-E Triad

The Langley-technique is the most popular procedure to estimate the extraterrestrial constant ETC. In practice, with respect to TOC measurements, the ETC can be calculated directly fitting a linear equation to the $MS9$ values respect to air mass μ:

$$MS9 = ETC + O_3 \alpha \mu \tag{6}$$

or using the Dobson method (Dobson and Normand, 1958; Komhyr et al., 1989):

$$\frac{MS9-(ETC+\Delta ETC)}{\mu}=O_3\alpha \qquad (7)$$

where Δ ETC represents the variation of this parameter respect to a reference value.

Both equations can be used to calculate the ETC value to be used in Eq.1 but Eq.7, where the slope is inverse to μ, has the advantage of presenting a better data distribution (Kiedron and Michalsky, 2016). This is because the number of measurements performed at low air mass ($1 \leq \mu \leq 2$) are more than those at large air mass ($\mu \geq 2$). Although all measurements could be used for this calculation, experience suggests that is better to select a subset of measurements. The days with a high aerosol optical depth concentration, i. e. during Saharan dust intrusions, are removed from this study with the help of data reported by other instruments.

The methodology used is essentially the same as described in (Redondas, 2008; Redondas et al., 2014b). The following criteria, listed in order of application, can be used to get a good agreement between the ETC values calculated:

1. The regression is performed on the [1.25 , 3.5] airmass range, using the brewer astronomical formulas for the airmass determination.

2. The data recorded during the morning and afternoon are taken separately (2 Langley per day)

3. Individual measurements (not the average of 5) are considered with the cloud screen method of 2.5 ozone standard deviation.

4. This daily standard deviation limit (2.5 DU) are used to select the Langley events.

5. MS9 double ratios are corrected for filter non linearity.

It is important to indicate that despite selecting the better days when the ETC values obtained for different days are compared, these present a standard deviation around $\pm$ 5. This difference is considered normal and the ETC introduced in Eq. 1 corresponds to the ETC mean. Although the median can be another option to get the ETC, the previous criteria guarantee that the difference between both methods are not significantly.

Aside from the interest to determine the TOC, the ETC is considered as a probe to check the correct state of the instrument. The ETC calculated from the Langley method presents a near constant value (std. $\pm$ 5), changing only when the instrument recalibrates. This may happen, for example, after replacing a damaged component or due to normal drifts by its continued operation. In both cases, and after a stabilization period, a new ETC value can be calculated.

As an example, Fig. 2 shows the operative value of the ETC for the Brewer #183 during the year 2011. The vertical lines represent situations which can produce a change in the behaviour of the instrument, while the horizontal line represents the operative ETC used to calculate the TOC (Eq.1). As it can be observed, the ETC changed twice, the first time by maintenance tasks (performed by IOS service in July 2011), and the second time due to changes in the Brewer configuration (to be more

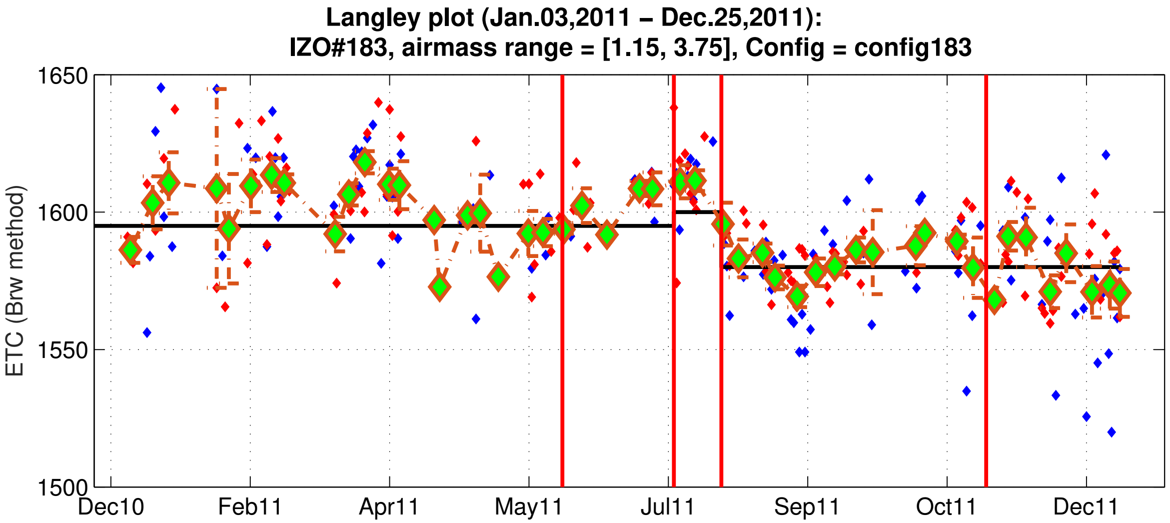

Figure 2. Operative ETC value for Brewer #183, after some events that could cause a change in the instrumental calibration during 2011. The red and blue dots denote daily ETC values calculated by the Langley method before and after solar noon, respectively. The diamond symbols and the error bars correspond to the ETC weekly mean and its standard deviation.

precise, changes in the so-called "Cal-Step" in August 2011). On the contrary, during the maintenance tasks (June 2011) or after UV calibration in our facilities (November 2011), the ETC remained constant. Only the Langleys that satisfy the conditions indicated in Sect. 2.2 are used to calculate the weekly mean. Other examples of events that may affect the ETC can be found in the calibration campaign reports (Redondas et al., 2015; Redondas and Rodriguez-Franco, 2016).

When a new ETC is given, the TOC calculated from it can be compared with the data obtained by other instrument with similar precision. This can be a strategy to check if the new ETC is correct. At the RBCC-E Triad, this task is simple because the Brewers are constantly compared to each other, allowing to identify the exact moment when a Brewer needs a new calibration. In addition, the traceability between the RBCC-E and the World Reference Triads is checked in the calibration campaigns, organized by the RBCC-E, comparing the data of the travelling references: Brewers #185 and #017. The results of these comparisons are shown in the calibration campaign reports (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2015; Redondas and Rodriguez-Franco, 2016).

3 Ozone and Dataset selected

In order to summarize the history of the RBCC-E Brewers, Table 1 provides the total number of days and measurements performed by these instruments since they became operational at IZO and as long as the weather conditions allowed them to

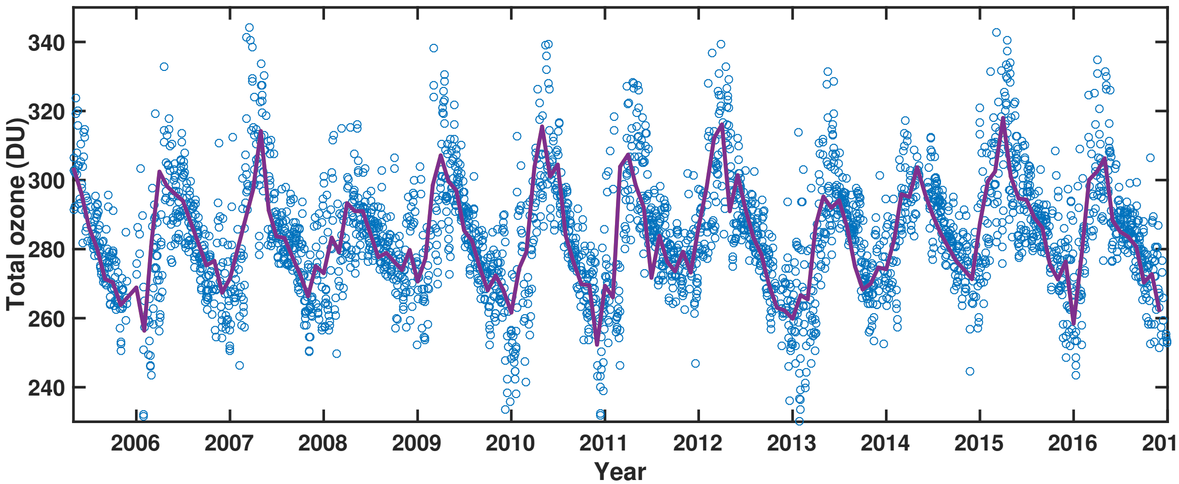

Figure 3. Time series of the ozone concentrations measured by Brewer #157 at Izaña Atmospheric Observatory, showing daily (dots) and monthly (line) means.

operate. Fig. 3 shows the daily and monthly TOC means measured by Brewer #157. The ozone presents an annual cycle with a sawtooth profile with maximum and minimum values in spring and autumn, respectively. Despite this annual behaviour, the ozone presents a lower daily variability as indicated by our measurements. This factor, together with a thermal inversion which produces an atmosphere free of anthropogenic pollutants and excellent weather conditions all the year, except for days with dust intrusions, explain why Izaña Atmospheric Observatory is an excellent location for a Brewer reference centre and, also, why the Langley-technique is used as calibration method.

In this work, the ozone datasets have been analyzed. The first dataset is obtained directly, after applying several conditions, listed next in order of application:

1. Only include measurements performed at Izaña Atmospheric Observatory.

2. Remove days with problems clearly identified (wrong alignment, etc.).

3. Only the days where the three Brewers have performed measurements are considered in this work. Moreover, each Brewer must have more than 12 measurements, with a minimum of 4 before and after the solar noon (homogeneous distribution).

The second dataset is obtained with the same conditions but also imposing the condition that the measurements must be simultaneous in time. A measurement made by a Brewer is considered simultaneous if in a temporal window of five minutes there is other measurement made by the other two brewers of the triad. Therefore, this second dataset can be considered a subset of the first. Table 2 gives a summary of both datasets. The entry “Evaluated Days” denotes the number of days used in each dataset to study the stability of the RBCC-E Triad. It is important to note that Dataset 1 includes the measurements made in the period 2005-2016 while Dataset 2 only considers simultaneous measurements from 2010 onwards. Before 2010, a large part of the measurement time was focused on the UV spectrum, and, hence, there are fewer ozone direct sun measurements in the instrument schedule. This means that the likelihood of finding 12 simultaneous measurements between the three instruments is low, particularly in winter where the presence of clouds is greater. After 2010, The RBCC-E started using the same synchronization schedule in their Brewers. These schedules take into account the sunrise and sunset times of each day and the routines, introduced in it, are distributed in function of the solar zenith angle (SZA).

Table 1. Number of operational days and measurements since their setup for the Brewers of the RBCC-E Triad.

	Brewer #157	Brewer #183	Brewer #185
Operational Days	3173	2740	2780
Operational Measurements	259534	204022	229201

Table 2. Summary of the datasets used in this work.

	Dataset 1	Dataset 2
Evaluated Days	2073	1325
Period studied	2005–2016	2010–2016

4 Results and discussion.

The stability of the measurements carried out by the RBCC-E Triad was evaluated from the datasets described in Sect. 3. In the case of the long-term behaviour, it was studied using both datasets, while the short-term behaviour was analyzed using only Dataset 1. The results obtained are shown in statistical terms.

4.1 Representative value of the Total ozone column.

To our knowledge, there are only a few publications where the stability of the World Reference and Arosa Triads are analyzed. In these articles, and due to the large number of ozone measurements performed each day, the authors have calculated a representative value of all of them and, from it, the long-term stability is analyzed (Fioletov et al., 2005; Scarnato et al., 2010; Stübi et al., 2017).

Fioletov et al. (2005) studied the long-term stability of the World Triad Reference in the period 1985 – 2003. In this work, the authors proposed to fit the measurements performed by each Brewer (#008, #014 and #015) to a 2nd grade polynomial (see Fig.4):

$$O_3 = A + B \cdot (t - t_0) + C \cdot (t - t_0)^2 \tag{8}$$

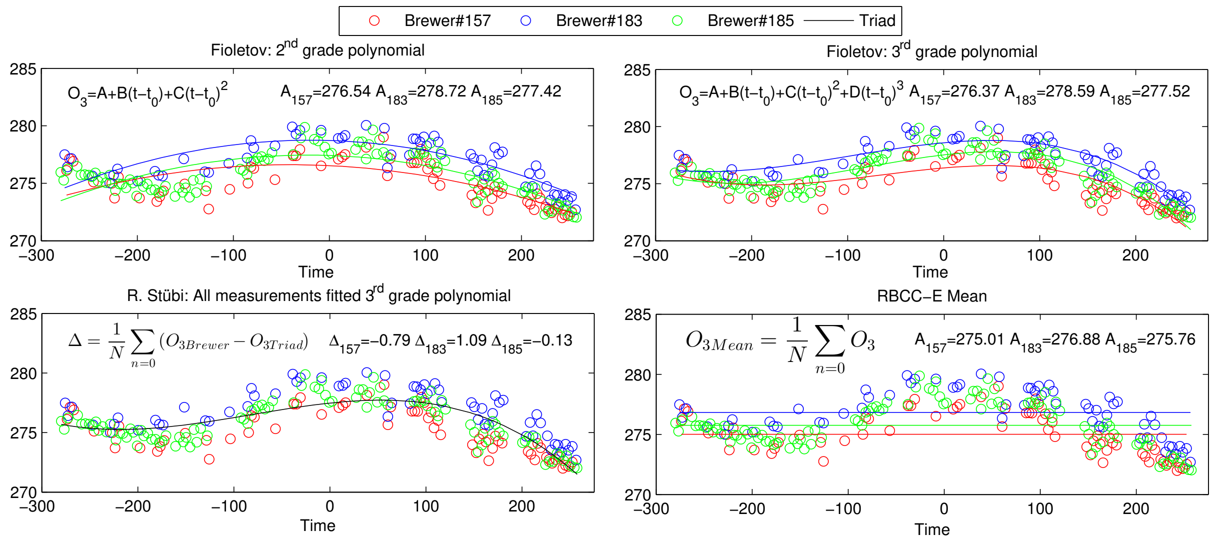

Figure 4. Method used to calculated the daily representative value. The ozone values plotted were measured the 16^{th} November 2016.

where O_3 is the TOC measured and $t - t_0$ corresponds to the difference between the time of the measurement and the solar noon. The independent coefficient A obtained through the adjustment is used as a representative ozone value of each instrument. The difference between this coefficient for each Brewer and the Triad mean represents the daily drifts of each instrument. The stability is studied from the relative standard deviation of these differences. In this work, the results using a 3$^{\text{rd}}$ grade polynomial have also been investigated, see Fig.4.

Stübi et al. (2017) studied the long-term stability of the Arosa Triad in the period 1988 – 2015. In this study, the authors considered that the behaviour of the Triad is the most appropriate reference for each day. Therefore, all the measurements made by the three Brewers are modeled as a 3$^{\text{rd}}$ grade polynomial dependent on time which represents to the Triad (see Fig.4):

$$O_3 = A + B\cdot(t - t_0) + C\cdot(t - t_0)^2 + D\cdot(t - t_0)^3 \quad (9)$$

where t_0 corresponds to the 12 UTC time. In this case, each Brewer can be characterized by a shift, Δ, which is the daily mean of the difference between the values measured and obtained from the fit and a standard deviation, σ, which evaluates the dispersion of these differences. Both parameters are used to analyze the long-term stability of the Arosa Triad.

In order to compare the long-term stability of the RBCC-E Triad with respect to the World Reference and Arosa Triads, both methods are used to fit our measurements. However, in this work, the time reference t_0 is the solar noon.

Although Eqs. 8 and 9 are valid to model the behaviour of ozone, it should be noted that the normal drifts by its continued operation of the instrument can affect the final value of the adjustment. Given this problem, calculating the daily mean of the measurements can be a good strategy to avoid this inconvenience. In this work, and noting by our measurements that ozone presents a reduced daily variability,see Fig.4, the mean of the all measurements, N, made by each Brewer was used as a representative value.

$$A_M = \frac{\sum_i O_3}{N} \quad (10)$$

The difference between the value obtained for each Brewer and the Triad mean represents the drifts of each instrument. Although the median can be another possibility to study the behaviour of the RBCC-E Triad, our experience suggests that the mean is robust. Moreover, since 2003, the mean has been used to detect when one of our Brewer loses its calibration, therefore, it has been interesting to include it in this work. At this point, it is important to note that for the World Triad Reference as for the RBCC-E, the representative value of each instrument is calculated, directly, from their measurements. In contrast, in the Arosa Triad, the representative value of each instrument (denoted as shift Δ) is calculated with respect to the behaviour of the three instruments, obtained by adjusting to a polynomial of the third degree (see Fig.4).

4.1.1 Long-term stability: daily averages

Following the procedure described by Fioletov et al. (2005) for the World Triad Reference, Datasets 1 and 2 (see Sect. 3) were fitted by a 2^{nd} and 3^{rd} grade polynomial. The distribution of the daily difference between the A value, see Eq.8, obtained for each Brewer with respect to the Triad mean are plotted in Fig. 5. Also, in this plot, the difference calculated from the daily mean of each instrument was included, see Eq.10.

It is important to take into account that the individual coefficients obtained for each Brewer, and also the Triad mean calculated from them, depend on the method used. As Fig.5 shows, the histograms that represent the results obtained after applying a polynomial fit are similar regardless of the dataset. This can be explained by the small daily variation of the ozone what causes the 2^{nd} and 3^{rd} grade polynomial fit to give very similar A coefficients, see Fig.4. In contrast, the histograms associated with the daily mean suggest that the differences between the brewers are less. This allows us to conclude that the method selected to evaluate the stability plays an important role, because the Brewer-Triad mean differences are directly associated with it. In this case, it may be more appropriate to use the daily mean to evaluate the RBCC-E Triad. Regardless, independently of the method used, Fig. 5 shows that, for the great majority of days, the Brewers present less than 2 DU of difference with respect to the Triad mean which indicates a good agreement among themselves.

Using the same procedure to evaluate the long-term stability can be the best strategy to compare different triads to each other. In this sense, Fioletov et al. (2005) only reported that the relative daily standard deviation of World triad reference is equal to 0.47%. This value was calculated as the mean of the relative standard deviation of each brewer. Table 3 contains the difference mean, calculated from the mean Brewer-Triad difference plotted in Fig. 5, and its standard deviation. The RBCC-E Triad presents a relative standard deviation mean equal to 0.41% ($\sigma_{157} = 0.362\%$, $\sigma_{183} = 0.453\%$ and $\sigma_{185} = 0.428\%$; see Table 3, Dataset 1, 4^{th} column). This result indicates that the dispersion of the measurements of the RBCC-E Brewers presents a similar behaviour to those of the World Triad Reference. Furthermore, the standard deviation values obtained confirm that the daily mean is the best method to evaluate the RBCC-E Triad.

In order to compare the daily behaviour of the Arosa and RBCC-E Triads, a 3^{rd} grade polynomial was fitted to all the daily measurements made by RBCC-E Brewers for Datasets 1 and 2. Then, for each Brewer its mean shift, Δ, and standard deviation,

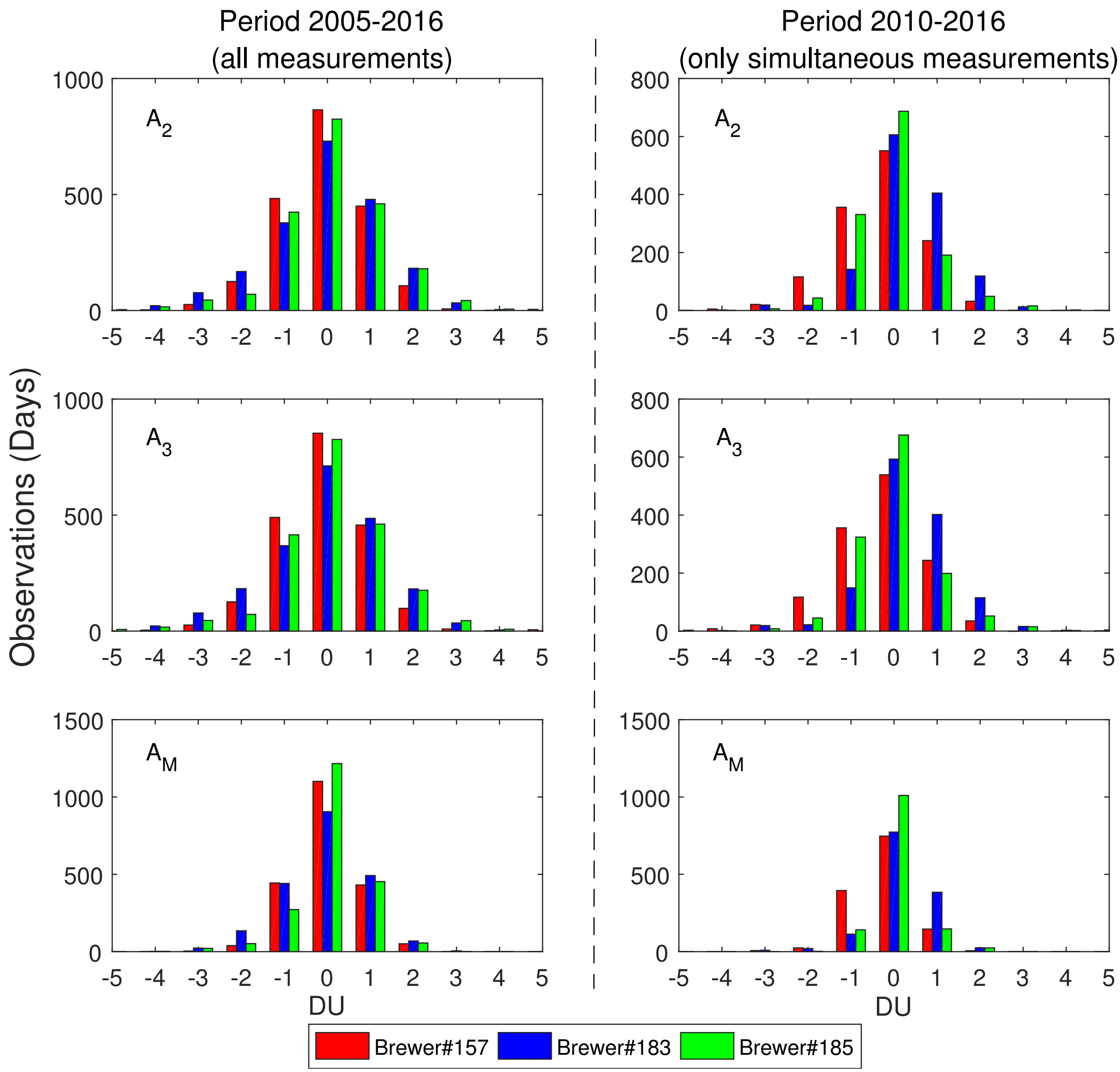

Figure 5. Daily difference of the ozone reference value A of each Brewer with respect to Triad. The values were obtained from the procedure proposed by Fioletov (World Triad Reference) and by daily mean (RBCC-E).

σ, were calculated (see Sect. 3). The values obtained for the Dataset 1 are shown in Fig. 6. Because Brewer #183 was damaged by a storm and was inoperative between December 2005 and September 2006, the data plotted in that period were calculated from measurements of Brewers #157 and #185 only. Similarly, when Brewer #185 is away from IZO in calibration campaigns, the values plotted correspond to Brewers #157 and #183. Note that although these data were introduced in Fig. 6 to avoid gaps

Table 3. Absolute and relative values of the mean shift and the standard deviation.

Dataset 1

	2^{nd} grade polynomial (DU)	3^{rd} grade polynomial (DU)	Brewer Mean (DU)	2^{nd} grade polynomial (%)	3^{rd} grade polynomial (%)	Brewer Mean (%)
Brewer #157	0.787 ± 1.04	0.796 ± 1.07	0.569 ± 0.744	0.276 ± 0.362	0.279 ± 0.372	0.1994 ± 0.26
Brewer #183	0.989 ± 1.29	1.01 ± 1.31	0.741 ± 0.956	0.349 ± 0.453	0.356 ± 0.463	0.26 ± 0.337
Brewer #185	0.89 ± 1.21	0.90 ± 1.23	0.56 ± 0.78	0.315 ± 0.428	0.32 ± 0.438	0.20 ± 0.279

Dataset 2

	2^{nd} grade polynomial (DU)	3^{rd} grade polynomial (DU)	Brewer Mean (DU)	2^{nd} grade polynomial (%)	3^{rd} grade polynomial (%)	Brewer Mean (%)
Brewer #157	0.795 ± 1.00	0.82 ± 1.05	0.534 ± 0.661	0.278 ± 0.349	0.286 ± 0.368	0.186 ± 0.23
Brewer #183	0.747 ± 0.942	0.784 ± 1.02	0.547 ± 0.719	0.262 ± 0.331	0.275 ± 0.36	0.192 ± 0.254
Brewer #185	0.64 ± 0.87	0.67 ± 0.99	0.376 ± 0.526	0.227 ± 0.311	0.238 ± 0.333	0.133 ± 0.189

in the plot, they are not considered in the statistical study. Therefore, the dates evaluated correspond with the days when the full RBCC-E Triad is operative, and the criteria established in Sect. 3 are still used.

As can be observed in Fig. 6, the results obtained for all instruments in Dataset 1 show a (± 0.5) value for the mean shift. A similar result was obtained for Dataset 2, figure not shown. Contrary to the report in Stübi et al. (2017), in the present case the standard deviation does not show any seasonal component. Again, this result is explained by the low daily variability of the ozone at sub-tropical latitudes. For the Brewers of the RBCC-E Triad, the standard deviation is more influenced by any anomalous internal behaviour of the instruments. For middle latitudes, e.g. in Arosa, there is a larger daily variation in ozone and the standard deviation shows it.

Following Stübi et al. (2017), Table 4 shows the distribution of percentiles of the mean shift and the standard deviation values plotted in Fig. 6. The Brewers present a similar interpercentile range $P_{2.5} - P_{97.5}$, with a mean value close to 1.1%. This result is consistent with the standard deviation shown in Table 3 for the polynomial fits. In comparison with the Arosa Triad, only Brewer #040 shows a better behaviour than the RBCC-E Brewers while their other Brewers (B#072, B#156) show similar values to ours.

4.1.2 Long-term stability: monthly averages

Although the histogram and the statistical parameters already presented suggest that the long-term stability of the RBCC-E, Arosa and World Reference Triads are similar. It can be more interesting to present this study from the monthly means. With this idea in mind, the daily difference plotted in Fig. 5 and the daily shift plotted in Fig.6 were monthly averaged. Fig.7 shows,

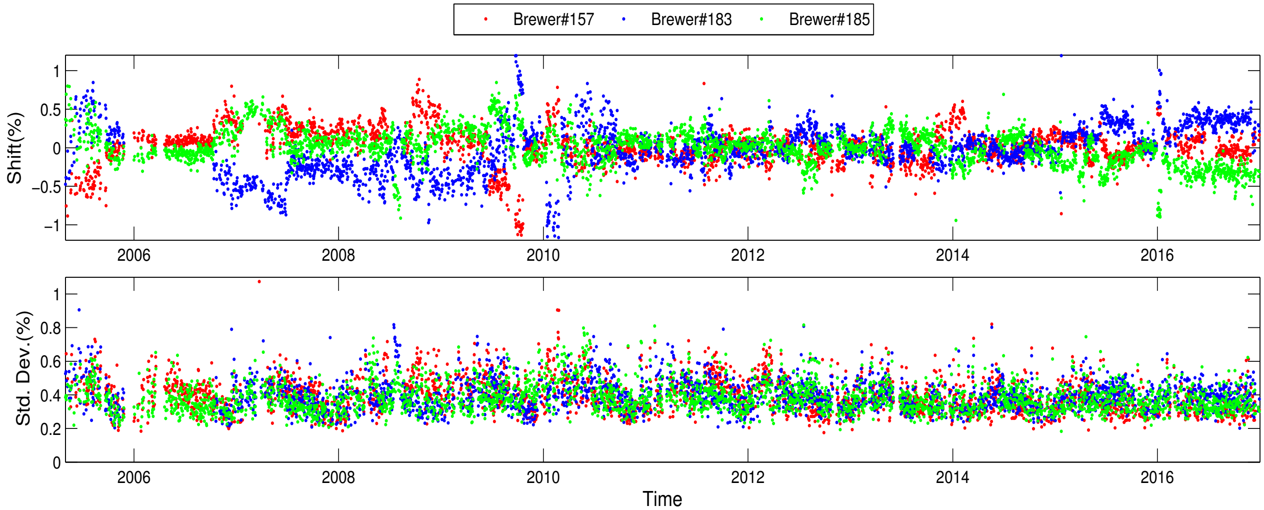

Figure 6. Time series of the mean shift Δ and the standard deviation σ, in terms relative to the TOC calculated from measurements performed by the three Brewers of the RBCC-E Triad (#157, #183 and #185) fitted with a 3[rd] grade polynomial

Table 4. Percentiles of the difference distribution (%) for the RBCC-E Triad.

Dataset 1			Shift Δ						Standard Deviation σ		
Brewer	$P_{2.5}$	P_{25}	Median	P_{75}	$P_{97.5}$	Brewer	$P_{2.5}$	P_{25}	Median	P_{75}	$P_{97.5}$
#157	-0.426	-0.13	0.017	0.138	0.464	#157	0.25	0.32	0.37	0.426	0.595
#183	-0.71	-0.219	-0.016	0.191	0.58	#183	0.25	0.327	0.377	0.439	0.599
#185	-0.54	-0.099	0.0155	0.147	0.51	#185	0.24	0.316	0.375	0.451	0.631
Triad	-0.599	-0.14	0.008	0.156	0.524	Triad	0.248	0.321	0.373	0.439	0.609

Dataset 2			Shift Δ						Standard Deviation σ		
Brewer	$P_{2.5}$	P_{25}	Median	P_{75}	$P_{97.5}$	Brewer	$P_{2.5}$	P_{25}	Median	P_{75}	$P_{97.5}$
#157	-0.573	-0.265	-0.053	0.084	0.365	#157	0.25	0.33	0.389	0.459	0.651
#183	-0.475	-0.093	-0.037	0.264	0.559	#183	0.265	0.349	0.405	0.473	0.627
#185	-0.341	-0.103	0.007	0.114	0.513	#185	0.247	0.33	0.389	0.479	0.6509
Triad	-0.503	-0.136	-0.001	0.1386	0.517	Triad	0.242	0.33	0.38	0.464	0.639

in relative terms, the monthly values for the period 2005–2016 (Dataset 1). The results confirm that the RBCC-E Triad has a good long-term precision, regardless of the method selected. In order to compare with the World Triad Reference, Table 5 contains the relative standard deviation of the difference between the representative value A of each brewer, calculated from the 2^{nd} grade polynomial fit, and the triad mean. As reported by Fioletov et al. (2005), the relative standard deviation of the

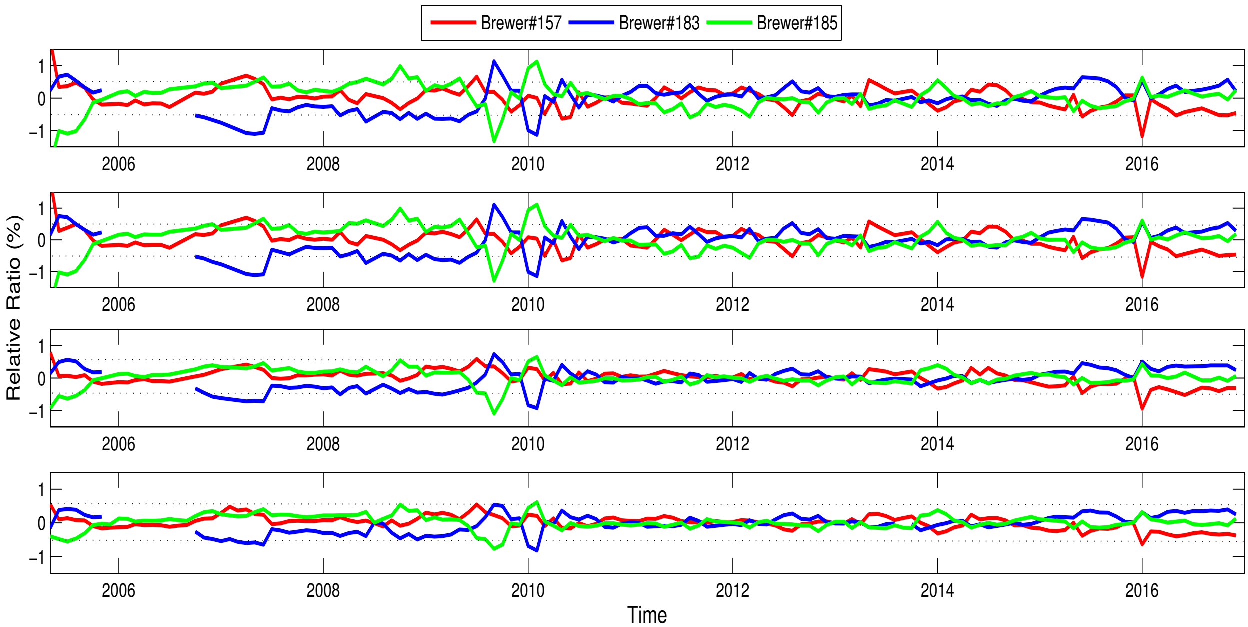

Figure 7. Relative ratio of the monthly values with respect to the Triad mean for the method proposed for World Triad Reference, Fioletov et al. (2005), Daily Mean (RBCC-E) and Arosa Triad, Stübi et al. (2017). The gap for the Brewer #183 data was caused by the tropical storm "Delta" which damaged the instrument. In 2010, the Brewer #183 had a problem with their micrometers

3-monthly mean for the World Triad Reference is 0.40%, 0.46% and 0.39% for Brewers #008, #014, and #015, respectively (0.42% in mean). The RBCC-E Brewers have lower 1-monthly and 3-monthly relative standard deviation. The ratio between 3-monthly values is 40% lower for the RBCC-E.

Table 5. RBCC-E and World Reference Triads: Relative monthly standard deviation.

	RBCC-E		WORLD REFERENCE	
	1-Monthly	3-Monthly		3-Monthly
Brewer #157	0.33%	0.29%	Brewer #008	0.40%
Brewer #183	0.34%	0.31%	Brewer #014	0.46%
Brewer #185	0.23%	0.20%	Brewer #015	0.39%
Mean	0.33%	0.27%		0.42%

Furthermore, Stübi et al. (2017) reported that in the period 2004–2012 the Arosa Triad presented monthly shift around at $\pm 0.4\%$ while the RBCC-E are lower than $\pm 0.3\%$.

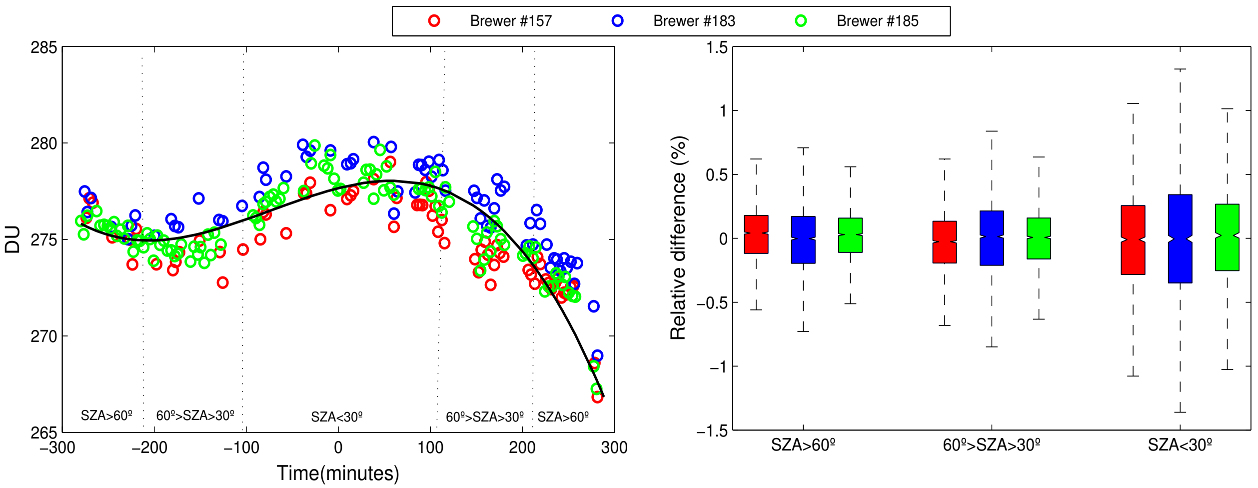

Figure 8. Experimental measurements and 3rd grade Triad fit (left) and relative difference by Brewer as function of the SZA (right).

4.2 Short-term stability

Dataset 1 was used to study the short-term stability of the RBCC-E Triad, with a view to determine in which SZA range the consistence of the measurements is higher. The measurements made by the three Brewers every day were fitted by 3rd grade polynomial as shown in Fig. 8. As previously commented, this polynomial represents the behaviour of the triad and allow obtain the TOC as a function of the time. Similarly to the previous study, each Brewer was characterized by a shift Δ. In this case, the data were divided as a function of the SZA. Different SZA ranges were checked, finding that the analysis can be reduced to just three broad ranges:

1. SZA>60º corresponds to the first and last ozone measurements of every day, when solar radiation presents a low intensity and high Rayleigh scattering.

2. SZA<30º corresponds to the measurement in the middle of the day, when the air mass is close to 1 and, hence, there is less Rayleigh scattering.

3. 60º<SZA<30º, the rest of ozone measurements.

In Fig. 8, the box-plot shows the statistical distribution of the mean shift calculated for the ranges selected. As can be observed, the greatest dispersion values are at low SZA which indicates at solar noon when more discrepancy can be observed between the data recorded by the instruments. This result may seem surprising because in these conditions the solar radiation on the Earth's surface is maximum and the Rayleigh scattering is minimum but it can be easily explained from Eq. 1. In this expression, at low SZA the optical mass, μ, is close to 1 and the ozone absorption α is a constant value. This implies that the

Table 6. Summary of the three studies comparing of the relative standard deviation of the World Triad Reference, Arosa and RBCC-E Triads

	daily	monthly	3-monthly
World Triad Reference	0.42	-	0.47
Arosa Triad	0.5	0.36	-
RBCC-E	0.41	0.37	0.27

The standard deviation calculated from procedure of Fioletov et al. (2005) and Stübi et al. (2017) are not equal but they are similar enough to compare the three triads. In this table, the values introduced for the RBCC-E were obtained from the procedure used by Fioletov et al. (2005) for the World Reference Triad.

denominator takes an almost constant value. Therefore, a small fluctuation (noise) associated with the MS9 values may affect significantly the TOC recorded. In contrast, for the other ranges the denominator takes a significant value and the effect of the noise on MS9 is less, increasing the stability of the measurements. In addition, the Fig. 8 shows that the Brewer #183 presents the poorer results which can be explained if it is taken into account that this instrument was damaged in 2007, and during 2008 it had an irregular operation. Moreover, in 2010, it had a problem with their micrometers (Roozendael et al., 2013).

5 Conclusions

The consistency of TOC measurements made by the RBCC-E Triad has been studied in order to check its long-term stability to compare it with the values reported for the World Reference and Arosa Triads. With this idea in mind, the procedures used by these triads were reproduced in this work to analyze the RBCC-E data. From the method used to evaluate the World Triad Reference, the difference between the measurements made by each Brewer and the Triad mean present a relative daily standard deviation equal to $\sigma_{157} = 0.362\%$, $\sigma_{183} = 0.453\%$ and $\sigma_{185} = 0.428\%$ (mean $\sigma = 0.41\%$), lower than the reported value for the World Triad Reference ($\sigma = 0.47\%$). Using monthly averages of these differences, the relative standard deviation reports values slightly lower than those obtained from the daily data. In addition, applying the procedure used to study the Arosa Triad, the RBCC-E Triad presents a similar interpercentile range $P_{2.5} - P_{97.5}$, with a value close to 1.1%, similar to those reported for the Arosa Triad, except in the case of Brewer #040. However, the monthly means are better for the RBCC-E Triad. Despite these differences, the values reported for each Triads are fairly similar, see Table 6, which ensures the traceability of the ozone measurements all around the world.

Competing interests. The authors declare that they have no conflict of interests.

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