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Dead time effect on the Brewer measurements: correction and estimated uncertainties

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Abstract

Brewer spectrophotometers are widely used instruments which perform spectral measurements of the direct and the global solar UV irradiance. By processing these measurements a variety of secondary products can be derived such as the total columns of ozone, sulfur dioxide and nitrogen dioxide, and aerosol optical properties. Estimating and limiting the uncertainties of the final products is of critical importance. High quality data have a lot of applications and can provide accurate estimations of trends. The dead time is characteristic for each instrument and non-proper correction of the raw data for its effect may lead to important errors in the final products. It may change with time and the currently used methodology is not always sufficient to accurately determine the correct dead time. For specific cases, such as for low ozone slant columns and high intensities of the direct solar irradiance, the error in the retrieved TOC, due a 10 ns change in the dead time from its nominal value, is found to be up to 5 %. The error in the calculation of UV irradiance is about 3–4 % near the maximum operational limit of light intensities. While in the existing documentation it is indicated that the dead time effects are important when the error in the used value is greater than 2 ns, we found that for single monochromator Brewers a 2 ns error in the dead time may lead to uncertainties above the limit of 1 % in the calculation of TOC; thus the tolerance limit should be lowered. A new routine for the determination of the dead time from direct solar irradiance measurements has been created and tested and a validation of the operational algorithm has been performed. Additionally, new methods for the estimation and the validation of the dead time have been developed and are analytically described. Therefore, the present study in addition to highlighting the importance of the dead time for the processing of Brewer datasets, also provide useful information for their quality control and re-evaluation.

1 Introduction

In the beginning of the 1980's, the increased concern for the stratospheric ozone depletion (Farman et al., 1985) and its effects on surface UV levels (Kerr and McElroy, 1993; Zerefos, 2002) stimulated the deployment of the first Brewer ozone spectrophotometers. Until 1996 Brewer instruments were manufactured by Sci-Tec Instruments Inc. at Canada. In 1996, Sci-Tec Instruments Inc. merged with Kipp and Zonen Inc. and since then they are produced at Delft, Holland. Nowadays, more than 200 instruments are deployed worldwide. Brewers are either single monochromators (versions MKII, MKIV, and MKV) or double monochromators (version MKIII) and are equipped with two types of electronics (old and new). Although of the same make, the characteristics of individual instruments may differ significantly. The Brewer network provides a variety of products such as the total columns of ozone (TOC) (Kerr et al., 1981), SO_2 (Cappellani and Bielli, 1995) and NO_2 (Cede et al., 2006; Diémoz et al., 2014), the aerosol optical depth (AOD) (Gröbner and Meleti, 2004; Bais et al., 2005; Meleti and Cappellani, 2000), as well as global and direct irradiance spectra (Bais et al., 1996, 1993). These measurements have supported scientific research for more than 30 years, enabling the investigation of their short-and long-term variability (Glandorf et al., 2005; Weatherhead et al., 1998; Zerefos, 2002) and interactions among them and among other atmospheric constituents (Bernhard et al., 2007). Additionally, good quality ground based measurements are very useful for the validation of satellite products which, under specific conditions, may be highly uncertain (Fioletov et al., 2002).

The uncertainty in the TOC retrieval is estimated to about 1 % (Kerr et al., 1985), while for well-maintained and properly calibrated instruments, the uncertainty of UV spectral irradiance is estimated to about 10 and 5 % for the UVB and the UVA spectral regions respectively (Bais et al., 1996). More recent studies indicated that the measurements can be largely affected by the individual characteristics of each instrument (Gröbner et al., 2006) and that proper corrections are needed in order to keep the uncertainties within the above mentioned limits, or even reduce them further. The non-

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ideal cosine response of the UV irradiance collector may lead to an underestimation of the diffuse component of the global irradiance by up to 12 % (Garane et al., 2006) and to an underestimation of the direct component that may exceed 20 % for solar zenith angles (SZAs) greater than 70° (Lakkala et al., 2008). The same studies suggest that the absolute response of the instruments may change by 0.2–0.3 % per 1 °C change of the internal temperature, depending on instrument and wavelength of the incident irradiance. Considering the TOC measurements from single monochromator Brewers, stray light effects can lead to underestimation of 2–6 % for ozone slant columns between 1600 and 2000 Dobson Units (DU) (Karppinen et al., 2014).

Yet, there are additional uncertainties related to constructional, technical or operational characteristics of the instruments, which are not adequately investigated and documented, and it is debatable whether the applied relevant corrections are optimal. The dead-time (DT) of the photon counting systems used in the Brewers is one of these characteristics. For about one third of the instruments participating in the RBCC-E calibration campaigns the actual DT derived differs from the DT used in the data correction by more than 2 ns, which is the maximum tolerable difference according to Granjar et al. (2008). Additionally, it is still not fully clear whether the current algorithm for the calculation of the DT and the correction of the data is the most appropriate (Redondas et al., 2012). Although there is some documentation for the theoretical description of the DT and for possible methods to determine the DT and correcting the data (Fountoulakis and Bais, 2014; Kerr, 2010; Kiedron, 2007; Kimlin et al., 2005; Redondas and Rodriguez, 2012; Rodriguez et al., 2014; Savastiouk, 2005; SCI-TEC Instruments Inc., 1999), there is little information regarding the associated uncertainties and for appropriate methodologies for correcting the measurements. The aim of the present study is to fill this gap in knowledge and to effectively contribute to the limitation of the uncertainties of the final products from Brewer instruments.

The objectives of this study have been addressed both experimentally and theoretically. Data from five different Brewers were processed and analyzed, specifically, from the double monochromator (type MKIII) Brewers with serial numbers 086 (B086), 157

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(B157), 183 (B183) and 185 (B185) and from the single monochromator (type MKII) Brewer with serial number 005 (B005). B005 and B086 operate at the Laboratory of Atmospheric Physics, Aristotle University of Thessaloniki, Greece (40.634° N, 22.956° E, 60 m a.s.l.). The Brewer instruments B157, B183 and B185 form the RBCC-E triad and are installed at the Izaña Atmospheric Research Center (28.309° N, 16.499° N, 2373 m a.s.l.). The same instruments were used in the closure experiments conducted for this study.

2 Dead time: calculation and correction of signal

2.1 The radiation detection system

The Brewer spectrophotometers use a photomultiplier (PMT) and a photon counting system for the detection and counting of the photons passing through the exit slit of the monochromator. A fraction of the photons that reach the PMT generate photon pulses, according to the quantum efficiency (QE) of the PMT (Haus, 2010), and are recorded as counts. The QE is a function of wavelength and is taken implicitly into account during the calibration. Each photon pulse has a finite temporal width and if two or more photons arrive within this time interval then they merge into a single pulse which is registered as a single count. This time interval is known as Dead Time (DT) and is characteristic for each instrument (Kerr, 2010). For most Brewer spectrophotometers the DT constant, i.e. the nominal value used for the correction of measurements, ranges between 15 and 45 ns. The DT effect increases non-linearly as a function of the rate of the incident photons (Kerr, 2010; Kipp & Zonen Inc., 2008; SCI-TEC Instruments Inc., 1999) and the correction of the measurements is complicated. During regular operation, the DT is calculated by measuring different levels of the irradiance emitted by an internal quartz-halogen 20 Watt lamp (standard lamp). The accuracy of the determined DT values depends strongly on the signal to noise ratio, thus on the level of the lamp's signal. A weak signal may lead to large uncertainties. Since the operation of the

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lamp depends on the operation of other electronic circuits in the instrument, it is not always easy to determine how much these factors may affect the calculated values of the DT. According to the manufacturer (Kipp & Zonen Inc., 2008; SCI-TEC Instruments Inc., 1999), the calculated values of the DT should not deviate from the nominal by more than 2 ns. However, during the regular operation of the instruments differences ranging from 2 to 10 ns are common (Redondas et al., 2012; Rodriguez et al., 2014). For B086 differences of up to 20 ns were found in the record of the monthly mean DT.

During the setup of a Brewer, the high voltage of the PMT is set to a value for which the dark signal is less than 100 counts s^{-1} when the signal of a measurement is about 10^6 counts s^{-1}. This is translated into a signal to noise ratio of $\sim$ 10 000 (Kipp & Zonen Inc., 2008), which, however, may gradually change with time to lower values. In this case a proper re-adjustment of the high voltage is necessary. Based on data from RBCC-E calibration campaigns (Redondas et al., 2012) we estimated that for most of the participated instruments the signal to noise ratio is above the suggested threshold. For low signal to noise ratios the response of the instrument is no longer linear and high uncertainties are induced in both the calculation of the DT and the correction of the signal. When the signal is too high, it cannot be easily corrected for the non-linear response of the instrument. Thus, during regular operation, proper attenuation filters are used to keep the signal below $\sim 1.75 \times 10^6$ counts s^{-1}. When the signal exceeds 7×10^6 counts s^{-1} the measurements are interrupted to avoid damage of the PMT.

The DT of a Brewer spectrophotometer is determined according to the following procedure: at the exit of the monochromator there are six exit slits through which the radiation dispersed by the monochromator is directed to the PMT. When the monochromator is set for an ozone measurement (“zero” position) (Kipp & Zonen Inc., 2008) the nominal wavelengths ($\lambda_{0\to5}$) corresponding to each slit are 303.2, 306.3, 310.1, 313.5, 316.8 and 320.1 nm respectively. Each exit slit can be opened individually, while the others are blocked, using a rotating mask which is synchronized with the photon counting system. There is one extra position (7) on the mask for which two slits (corresponding to λ_2 and λ_4) are opened simultaneously. In order to determine the DT,

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the spectral irradiance of the standard lamp at 306.3 and 313.5 nm is measured by setting the rotating mask at positions 3 and 5 respectively, followed by a simultaneous measurement at both wavelengths by setting the mask to position 7. This sequence is repeated 10 times (10 cycles) and the DT is calculated by the methodology that is described in the following paragraph. The same procedure is repeated 5 times for high intensity signal and 10 times for low intensity signal, and then the mean value and the standard deviation for each set of measurements is determined. Two filter-wheels are interposed between the entrance of the optics and the PMT (Kipp & Zonen Inc., 2008), which will be referred as filter-wheel 1 (FW#1) and filter-wheel 2 (FW#2). Each wheel has six holes spaced at 60° intervals. One of the holes in each filter-wheel is empty while filters with different transmittance are placed in each one of the five remaining positions. Each hole can be selected to intersect the optical axis by rotating its filter-wheel. During the DT measurements, a quartz diffuser is selected for FW#1, while for FW#2 an empty aperture is selected for the high intensity measurements and a neutral density (ND) filter of optical thickness 0.5 (∼ 0.316 transmittance) for the low intensity measurements.

2.2 Theoretical approach of dead time determination

For a mean rate N of photons that reach a detector, individual measurements may differ from each other due to the quantized nature of light and the independence of photon detections (Hasinoff, 2014). Photon counting is a classic Poisson process and the Poisson (photon) noise of the measurements is reduced as the sampling time increases. The fractional 1-sigma precision ($\Delta S/S$) is given by the reciprocal of the square route of the photons measured within the time t:

$$\frac{\Delta S}{S} = \frac{1}{\sqrt{Nt}} \tag{1}$$

Due to DT loss of photons, the Brewer measurements have no longer Poisson distribution. Thus, Eq. (1) underestimates the 1-sigma precision and should be replaced by

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the more precise Eq. (2) which takes into account the DT effect (Kiedron, 2007):

$$\frac{\Delta S}{S} = \sqrt{\frac{1}{N_\mathrm{M} \cdot t} - \frac{\tau}{t} \cdot (2 - \frac{\tau}{t}) \cdot (1 - \tau \cdot N_\mathrm{I})^{-1}} \tag{2}$$

Where N_M is the rate of detected photons (counts), t is the total time of measurements, τ is the DT and $\mathrm{N_I}$ is the rate of incoming photons. As long as $\mathrm{N_I}$ remains below $\sim 3 \times 10^6$ photons s^{-1}, the results from Eqs. (1) and (2) do not differ by more than 2 % (Kiedron, 2007).

The algorithms which have been developed for the calculation of the dead time and for the correction of the signal for its effect are both based on Poisson statistics. According to Schätzel (1986), the average number of photons within the dead time τ for a mean rate of $\mathrm{N_I}$ pulses per second is given by:

$$\mu = \mathrm{N_I} \cdot \tau \tag{3}$$

For a Poisson distribution the probability $P(k)$ of k pulses within τ is then given by:

$$P(k) = \frac{1}{k!} \cdot e^{-\mu} \cdot \mu^k \tag{4}$$

The sum of probabilities for $k = 0$ to infinity should be equal to unity; thus the probability of exactly one pulse within τ is given by:

$$P(k=1) = \frac{1}{1!} \cdot e^{-\mu} \cdot \mu^1 = e^{-\mu} \tag{5}$$

while the probability for one or more photons within τ is given by:

$$P(k \geq 1) = 1 - P(k=0) = 1 - e^{-\mu} \tag{6}$$

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The ratio of the detected pulse rate (N_M) against the overall number of pulses (N_I) using Eqs. (5) and (6) is then:

$$R = \frac{N_I}{N_M} = \frac{P(k=1)}{P(k \geq 1)} = \frac{\mu \cdot e^{-\mu}}{1-e^{-\mu}} \approx \frac{\mu \cdot e^{-\mu}}{1-[1-\mu+\frac{\mu^2}{2}-\ldots]} \approx e^{-\mu} \tag{7}$$

By replacing μ from Eq. (3), Eq. (7) can be written as:

$$\frac{N_M}{N_I} = e^{-N_I \cdot \tau} \tag{8}$$

In the Brewer software, Eq. (8) is applied separately to the count rates for λ_2 and λ_4 (slit mask positions 3 and 5) and to count rates for λ_2 and λ_4 simultaneously (slit mask position 7), by setting $N_I^0 = N_M$ as an initial guess and then by iterating over the rearranged expression:

$$N_{Ii}^{j+1} = N_{Mi} \cdot e^{N_{Ii}^j \cdot \tau} \tag{9}$$

Finally, τ is determined by:

$$\tau^j = \frac{1}{N_{I7}^j} \cdot \ln(\frac{N_{I7}^j}{N_{M7}}), \text{ with } N_{I7}^j = N_{M3}^j + N_{M5}^j \tag{10}$$

After 9 iterations of Eq. (10) τ converges to a value that defines DT and is used for the correction of the measurements for the effect of DT, from Eq. (11):

$$N_I^{j+1} = N_M \cdot e^{N_I^j \cdot \tau} \tag{11}$$

Again, after 9 iterations the result converges to the corrected value of the count rate N_I^9.

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Questions have been raised (Kiedron, 2007) whether the algorithms used for the calculation of the DT and the correction of the signal are the most appropriate and whether the simplifications in the algorithm used for the DT calculation can lead to systematic underestimation of its value and subsequently of the corrected signal. These issues are addressed below.

Given the Poisson nature of photon statistics, there are two formulas that are commonly used to calculate the DT (Schätzel, 1986; Yu and Fessler, 2000), which depend on the nature of the counting system. For Brewers, the algorithm for the calculation of the DT has been based on the assumption that all photons, either recorded or not recorded by the PMT, trigger a new DT period (paralyzable system) and the extended DT formula is used (Eq. 8). If we assume that DT is triggered only from the photons that are recorded from the PMT (non-paralyzable system) then the non-extended DT formula should be used:

$$R = \frac{N_{\mathrm{I}}}{1 + \tau \cdot N_{\mathrm{M}}} \tag{12}$$

Equation (12) is derived from Eq. (8) by assuming a very small dead time and by replacing the exponential term with its Taylor expansion. Subsequently, a new equation corresponding to Eq. (11) can be derived:

$$N_{\mathrm{I}}^{j+1} = N_{\mathrm{M}} \cdot (1 + \tau \cdot \mathrm{N}_{\mathrm{I}}^{j}) \tag{13}$$

It is not clear if the formula used for the calculation of the DT in Brewers and for the correction of the measured signal is the most appropriate (Kiedron, 2007). Additionally, the simplifications applied in the algorithm for the DT calculation could lead to systematic underestimation of its value and subsequently to underestimation of the corrected signal. These concerns are addressed in the following.

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2.3 Experimental evaluation of DT determination

2.3.1 Extended and non-extended DT

The DT for five different Brewers was calculated by the two different approaches (expressed by Eqs. 11 and 13), in order to assess the resulting differences. The results are presented in Fig. 1. For both cases the calculated DT values were found to converge simultaneously after 9 iterations (Fig. 1a), while the differences between the derived DT values are negligible. Specifically, when the non-extended DT formula (Eq. 13) is used the calculated DT is less than 0.5 ns lower than for the extended DT formula (Eq. 11). In order to estimate the differences between the final products for a paralyzable and a non-paralyzable system, count rates from 0 to $10^{6.5}$ counts s^{-1} were assumed and corrected using both formulas for DT nominal values ranging from 15 to 45 ns (Fig. 1b and c).

We notice that in all cases the results are already converging after 4 or 5 iterations, simultaneously for both methodologies. This implies that it might not be necessary to perform 9 iterations for the correction of the signal. For count rates lower than 10^6 counts s^{-1} the differences between the corrected values with the two methods are lower than 0.1 %. For count rates between 10^6 and $10^{6.5}$ (~ 3.2 million) counts s^{-1} the differences become more important. For DT values below 30 ns the differences remain below 1 % for the entire range of count rates, while for DT of 45 ns, the differences exceed 1 % only for count rates higher than $10^{6.4}$ (~ 2.5 million) counts s^{-1}.

2.3.2 Artificial biases

In order to determine the conditions under which the standard Brewer algorithm does not induce artificial biases in the results the following procedure was followed: theoretical values of the measured count rates N_M were estimated for different incoming photon rates N_I and for different reference DT values by using Eq. (8). Then the DT was recalculated from Eqs. (9)–(11). As long as the ratio between the count rates at positions 3 (or 5) and 7 of the slit mask N3/N7 (or N5/N7) remains between 0.25 and 0.5 and the true count rate at position 7 (N7) remains below 10^7 counts s^{-1} (the maximum measured signal is usually below 3×10^6 counts s^{-1}), the calculated and the reference DT coincide.

On the other hand, as deduced from Fig. 2, the calculated DT is lower than the "real" DT when the ratio N3/N7 is below ~ 0.25 for all reference DTs. This result remains valid also for pulse rates different than 10^6 counts s^{-1}.

2.3.3 Dark signal

The thermal noise of the electronics is responsible for the dark signal which is recorded even when no radiation reaches the PMT. Thus in the Brewer algorithm, prior to the dead time correction, the dark signal is subtracted from the measured signal (Kerr, 2010). However, Kiedron (2007) suggested that before subtracting the dark signal both the measured and the dark signals should first be corrected for the dead time effect. In the same study it was suggested that even though the dark pulses have no Poisson distribution, using Eq. (12) for the correction of both the dark and the measured signals should lead to more accurate signal correction than if the dark signal is not corrected for the DT effect. In the following we attempt to quantify the differences in the final results between the two approaches concerning the correction of the dark signal for the DT effect. To achieve that, different levels of count rates, ranging from 0 to $10^{6.5}$ counts s^{-1}, were assumed. On these count rates different dark signals were added, ranging from 0 to 10^5 counts s^{-1}. Then the derived count rates were corrected for the dead time and the dark signal using both methods (operational in Brewers and suggested by Kiedron, 2007) and the resulting corrected signals were compared. For low dark signals (< 1000 counts s^{-1}) no differences could be detected. These are becoming more important as the assumed signal, the dark signal, and the dead time are increasing. However, as long as the dark signal is below 10^4 counts s^{-1}, the difference between the results from the two methods is lower than 0.2 %, even for a dead time of 60 ns. Since during normal operation of a Brewer the dark signal is generally lower than this level,

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the correction of the dark signal for the DT effect would not have important impact on the final results. The difference increases fast when the dark signal approaches 10^5 counts s^{-1}, but stays below $\sim 1.6\,\%$ even for count rates of $10^{6.5}$ counts s^{-1} and dead time of 60 ns.

2.3.4 Simplifications in the algorithm

In order to detect if the calculated DT is underestimated due to the simplifications in Eq. (7) or if the algorithm is not proper for reasons that have not been taken into account, the below described method has been developed.

Measurements of spectral irradiance emitted by 3 different sources (the sun, an external 1000 Watt DXW lamp and the internal 50 W standard lamp) were performed in steps of 5 nm for the operational spectral range of Brewer spectrophotometers using different ND filters (different positions of FW#2) and different levels of intensities. In order to achieve different levels of irradiance when the Sun is used as a radiation source, the measurements were performed for several solar zenith angles. Measurements of the 1000 Watt lamp intensity (DXW lamp with serial number 1005) were performed at the Izaña Atmospheric Research Center for different distances (ranging from 40 to 115 cm) between the lamp and the center of the quartz window of the Brewer. The lamp mount could move vertically on a metal rod of 1 m length. The intensity and the voltage of the lamp were continuously monitored in order to ensure that the lamp irradiance was stable during the measurements. Then the relative attenuation between the different positions of FW#2 is calculated (Sellitto et al., 2006; Redondas et al., 2011).

The measured spectral irradiance from all the different light sources were corrected for the DT using several values ranging from zero to about twice the used DT constant in steps of 0.1 ns. Then, the signal ratios (relative attenuation) between all the different positions of FW#2 were calculated for each wavelength. The transmittance of the ND filters is known to be independent of intensity. Assuming that the response of each instrument is non-linear (intensity dependent) exclusively due to DT, correction of the signal with the proper DT value (and method) should eliminate the non-linearity. For all

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wavelengths, the optimal DT correction should lead to signal ratios of pairs of FW#2 positions that are independent of the intensity of incident radiation. This also suggests that using an improper DT for correcting measurements for the determination of the attenuation of ND filters might lead to important errors. An example for one pair of ND filters and one wavelength is shown for B185 in Fig. 3. From Fig. 3a, the optimum DT for which the calculated attenuation is independent of the measured count rate is found at 29 ns, while lower and higher DT values result in remarkable differences.

The above described procedure was repeated for all wavelengths and for all possible combinations of filters. Irradiance values for which the noise to signal ratio is very high were not used. Additionally, some outliers resulting from spikes (Meinander et al., 2003) were rejected by visual inspection. The mean DT and the corresponding standard deviation are then calculated from the remaining values, as shown in Fig. 3b.

For B185, the optimum correction DT is very close to both the nominal value and the mean DT calculated from the standard lamp measurements. The standard deviation is nearly 2 ns. The same test was performed for B086, operating at Thessaloniki. In this case moving the 1000 Watt lamp vertically was impossible; thus the lamp was fixed to a standard distance of about 40 cm from the center of the quartz window and different intensities were achieved by adjusting the lamp current. For B086 the test was performed for two periods with different calculated mean DT. In both cases the results were within 1 ns from the mean DT calculated with the standard procedure, and the standard deviation was again of the order of 2 ns. The test is more uncertain when applied on single-monochromator Brewers, mainly due to the stray-light effect.

The results discussed above reveal that the algorithm currently used is reliable and provides an accurate estimation of the DT, as long as the count rates at positions 3 and 5 of the slit mask do not differ significantly. However, as it is analytically explained in the following paragraphs, operational or technical issues of the instrument may affect the determination of the DT, leading to important errors in the measured signals, and consequently, in the derived final products.

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2.4 Determination of DT from solar measurements

Using the standard lamp as a radiation source for the determination of the DT may lead to uncertain and noisy results, especially when the signal of the lamp (thus the signal to noise ratio) is low. Such results, in addition to inducing errors in the proper correction of the measured signals, also make difficult the detection of possible problems (of mechanical or electronic origin) that may affect the DT. The operation of the lamp is not independent of the operation of the rest electronic circuits of the instrument. Thus, it is not always easy to detect if observed changes in DT are real. The sun is a more reliable and stable (under specific conditions) source compared to the standard lamp; thus using the solar measurements for the calculation of the DT would eliminate a great part of the uncertainties. New routines for the determination of the DT from direct sun measurements were created and tested on Brewers 005, 086, 157, 183, and 185 during a period of about 10 months.

The methodology used for the DT determination from direct sun measurements is very similar to that described in Sect. 2.1. The main differences are in the measurement procedure: the zenith prism points towards the sun instead to the internal lamp, and appropriate ND filters are used in order to avoid PMT overexposure. Different number of cycles (different signal integration times), ranging from 10 to 40 (integration time of ~ 1 to ~ 4.5 s), was used in different instruments during different periods. At Thessaloniki, five consecutive measurements were performed each time and then the mean dead time and the standard deviation were derived. The gratings were moved to a position where the N3/N7 ratio remained within the acceptable limits, as discussed previously. At Izaña, the mean dead time and the standard deviation were derived from four consecutive measurements, with the gratings set at the ozone measurement position. The main problem when using the sun as radiation source is the possibility of being partially or fully covered by clouds, resulting to rapidly changing or very low irradiance, respectively. In these cases, the uncertainty of the calculated DT is extremely high. Thus, in this analysis we rejected all measurements with standard deviation higher than 5 ns

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and with count rates at position 7 of the slit mask below 10^5 counts s^{-1}. In Fig. 4, the DT derived for three of the five Brewers studied are presented as a function of day of the year (DOY).

For B086 the DT derived from the standard lamp is much noisier than from the sun during the first months of the year. This is due to the very low intensity of the standard lamp used in that period. In April (DOY 94) the standard lamp was replaced with one of higher intensity. Accordingly, the noise in DT results was reduced. Further improvement in the DT results can be observed after DOY 142, when the number of cycles was increased from 10 to 20. During the analyzed period, the DT as calculated from measurements of solar radiation is very stable and less noisy than the DT from the standard lamp. Prior to August 2014 (DOY 220) the direct sun measurements for the determination of the DT were performed only once per day near the local noon in order to have very stable solar irradiance with high intensities. Although the noise is very low during this period, there are too few measurements available. Since the beginning of August, several measurements were performed per day at different solar zenith angles; thus the amount of available data has increased, but also the level of the noise. B086 is a very insensitive instrument; therefore any reduction in the level of intensity is reflected immediately on the estimated DT.

For B157, the agreement between the DT from the standard lamp and the sun is very good. Some outliers in DT derived from the sun measurements are due to the inclusion data recorded at large solar zenith angles when the intensity was low. It should be noted that in this case, as well as in the case of B086, the calculated DT (from both the internal standard lamp and the sun) is lower than the nominal. Considering that B157 is a well maintained instrument, the DT used for the signal correction should be 4–5 ns lower than the used DT constant of 32 ns, at least for the presented period.

For B185, the DT from the sun is lower and noisier compared to the DT from the standard lamp. The main reason is the low ratio of the count rates between positions 3 and 7 of the slit mask when the DT is calculated from direct sun measurements. The ratio ranges between 0.05 and 0.25 for the majority of the direct sun measurements,

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while it remains within 0.4 and 0.5 when the irradiance of the standard lamp is measured. For the first part of 2014 (before DOY 190), the measured DT is lower than the DT constant used during this period. In this day the DT constant was changed from 33 to 29 ns, after ensuring that no realignment of the optics and no resetting of the PMT were needed, following the suggestion of Kimlin et al. (2005). The conclusions from the analysis of the DT for B086, B157 and B185 are valid also for B005 and B183; thus their results are not presented.

Rodriguez et al. (2014) suggested that the calculated dead time is not necessarily the one which provides the optimal signal correction and further investigation is needed. The present study showed that the procedure used for the determination of the DT provides accurate results as long as the measurements are performed under appropriate conditions (as already discussed in Sects. 2.1–2.3 and is further discussed in Sect. 2.5). However, even if the correct DT of the PMT is known and used, there are other factors that might lead to non-linear responses of the photon detection system which might be falsely perceived as improper DT correction.

2.5 Factors affecting the determination of DT

In order to determine the optimum instrumental settings for the calculation of the DT, continuous direct sun measurements were performed during two consecutive very clear days at Izaña with B185. About every 40 min, five consecutive DT measurements were performed using different grating settings, so that each time the irradiance passing through the two exit slits was of different wavelength. The five wavelengths corresponding to position 3 of the slit mask were 306.3, 317, 331.5, 345 and 354.5 nm, while for position 5 the five wavelengths were higher by about 7 nm. This way, measurements for different intensities, wavelengths, and N3/N7 ratios were performed for very similar SZAs and atmospheric conditions. During the first day, 40 cycles were used for the first set of wavelengths (306.3 nm for position 3) and 10 cycles for the other four sets. During the second day, the number of cycles was changed to 10 and 5 respectively.

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Due to the different combinations of measurements the recorded N3/N7 ratio ranges from 0.05 to 0.5. The DT derived from measurements with N3/N7 ratios between 0.3 and 0.5 is very close to the DT constant (29 ns). For the same value the optimum DT correction was achieved, as discussed in Sect. 2.3. As expected, for ratios lower than 0.3 the DT is underestimated. The irradiance level does not affect the mean calculated DT; but is important for the uncertainty of the measurements. The calculated standard deviation decreases with increasing count rates. For count rates (at position 7) between 500 000 and 1 000 000 counts s^{-1} the standard deviation is smaller than $\sim 5\,\%$ (~ 2 ns) of the calculated DT, as long as the N3/N7 ratio remains within the acceptable limits of 0.3–0.5 and when 10 or more cycles are used in the measurements. For count rates higher than 10^6 counts s^{-1} the standard deviation is smaller than 3 % (~ 1 ns) of the calculated DT and decreases even more for higher count rates. Increasing the number of cycles also decreases the standard deviation and therefore the uncertainty in the determination of the DT. When the number of cycles increases from 5 to 10 the standard deviation decreases by a factor of 2. The same fractional decrease in the standard deviation is found also when the number of cycles increases from 10 to 40. It should be mentioned that no wavelength dependence was detected in the determination of the DT. The same occurs for the temperature effects. In order to look for possible dependencies from temperature, finally, the DT was calculated for different temperatures within a specific day by using measurements with the standard lamp and no change was detected for temperatures ranging from 17 to 35 °C.

3 Uncertainties of the final products

In the following, an attempt is made to quantify the main uncertainties in the calculation of UV irradiance, TOC and AOD due to the uncertainties in the estimation of the DT. Effects in the calculation of the total columns of SO_2 and NO_2 are not discussed, because uncertainties from other sources are much higher, due to the usually small column amounts (the order of a few DU) of these species (Fioletov et al., 1998;

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Dead time effect on the Brewer measurements

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Wenig et al., 2008). Dead time errors are also expected to affect the results of different diagnostic tests, such as the measured intensity of the internal lamps and the determination of the attenuation of the ND filters, and in turn the accuracy of the final products. It is difficult to quantify these uncertainties; however, they are believed to be of less importance compared to those discussed below.

3.1 UV irradiance

The effect of DT errors on the calculated UV irradiance depends mainly on the intensity of the incident radiation. Figure 5 shows the effect on irradiance caused by deviations in the DT, in the range ±2 to ±10 ns, from four characteristic reference values, as a function of the intensity (photons s^{-1}) of the incident radiation. As long as the intensity remains below 10^6 photons s^{-1}, even a large change of 10 ns in the DT leads to a corresponding change in the calculated irradiance of up to about 1 %. For higher intensities the effect increases rapidly, so that for intensities near $10^{6.5}$ (~ 3.2 million photons s^{-1}) a change in the DT of only 2 ns – a level that is usually encountered in Brewers – causes 1 % change in irradiance. Larger deviations, however, result in larger errors in irradiance that cannot be neglected. For example, a 10 ns change in the DT leads to differences in the calculated irradiance ranging from about 3 to 5 %, depending on the actual value of the DT.

In order to get quantitative estimates of the errors that may be introduced in the global spectral irradiance measurements due to uncertainties in the determination of the DT, data from Brewer instruments operating in Thessaloniki and Izaña were processed using different values of the DT. For instruments with low sensitivity, such as B086, a large change in the DT of 10 ns, relative to the nominal value of 42 ns, leads to a change in the noon global spectral irradiance of less than 2 %, even at UV-A wavelengths (strong radiation) during a very clear day near the summer solstice (~ 17° solar zenith angle at Thessaloniki). The same change in the DT for an instrument with high sensitivity, such as B185 (with a DT constant of 29 ns), leads to 4 % change in irradiance for local noon at Izana (~ 6° solar zenith angle), and about 2.5 % change in the

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daily integral of the irradiance at 350 nm. For smaller, and more usual, changes in the DT of 2 ns, the corresponding change in irradiance at local noon is less than 1 % while for the daily integral the change is negligible. Finally, when no correction for the DT is applied, the global irradiance is underestimated by up to 9 %. For wavelengths in the UV-B region, the effect of the DT is negligible since the radiation is much weaker for both instruments.

3.2 Total ozone column

The retrieval of TOC with a Brewer spectrophotometer is based on the analysis of near-simultaneous direct-sun spectral irradiance measurements at four wavelengths (Kerr et al., 1981). Five sets of measurements are performed within about 2 min and the mean TOC and the corresponding standard deviation are calculated. Before each set of measurements, the intensity of the irradiance is tested and an appropriate ND filter is inserted in the radiation path to avoid overexposure of the PMT. For the retrieval of TOC from direct sun measurements it is also necessary to know the extraterrestrial constant (ETC). The ETC can be either calculated by using the Langley extrapolation method (Thomason et al., 1983) or transferred from a standard instrument through side-by-side ozone measurements (Fioletov et al., 2005).

Because the correction for the DT applies to measurements of irradiance and its effect depends on the level of irradiance, the effect on the retrieval of TOC depends basically on the differences in the count rates at different slits (310.1, 313.5, 316.8 and 320.1 nm). Such differences are caused by atmospheric influences on the solar spectrum (e.g., from ozone absorption, Rayleigh scattering, and SZA) and by the shape of the spectral response of the instrument. The latter may significantly differ between instruments, particularly for Brewers of different type. For example, the presence of the $NiSO_4$ filter in single-monochromator Brewers changes significantly the shape of the spectral response, compared to double-monochromator Brewers, leading to different correlation between the levels of irradiance measured at the four slits. Although the shape of the spectral response differs between instruments which are equipped

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with different electronics the differences were not found to be as important as between single- and double-monochromator Brewers. In the following, the effect of the DT correction on the determination of the ETC and the TOC from direct-sun measurements are discussed for the single-monochromator B005 and the double-monochromator B185. The same analysis for the MKIII Brewers 157 and 183, not shown here, yielded similar results to those for B185.

3.2.1 ETC from Langley plots

Usually, in order to derive the ETC from Langley plots, continuous measurements of the irradiance for the wavelengths used for the calculation of TOC are performed during half day (morning to noon or noon to evening) with stable atmospheric conditions (clear sky, stable TOC, low and stable AOD). Then the ratio used for the calculation of TOC (Kerr et al., 1981) is derived and plotted against the air mass. The ETC is equal to the intercept of the resulting linear fit. Errors in the determination of the DT may introduce errors in the calculation of the ETC. Although the irradiance levels increase with decreasing SZA, the use of the ND filters prevents the PMT from exposure to very high intensities for which the role of DT is critical. However, the effect of the DT errors remains important when the signal is near its high intensity threshold. Langley plots for about 10 days were derived from measurements with the MKIII Brewers 157, 183, and 185 in Izaña and the MKII Brewer B005 in Thessaloniki. Although the atmospheric conditions at Thessaloniki are not usually favourable for the determination of the ETC with the Langley method, days with relatively stable atmospheric conditions were found within one year of measurements which were used indicatively for the purposes of this study. For the MKIII Brewers, the change in the derived ETC for a 2 ns change in the DT is typically lower than 3 units, rising up to $\sim$ 15 units for 10 ns change in the DT. The corresponding changes in the ETC for the MKII Brewer are 8 and 40 units respectively. Such errors in the determination of the ETC influence directly the calculated TOC. The reasons for the differences between the results for the two instruments are discussed in the following section.

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In Fig. 6, changes in the calculated TOC due to changes in the ETC resulting from typical errors in the DT are presented. The error in TOC increases smoothly with decreasing ozone slant column. For B185, the change in TOC due to a 2 ns change in the DT is generally less than 0.5 %, rising to about 1.5 % for a 10 ns change in the DT for slant ozone columns lower than 500 DU. For B005, the change in TOC for a 2 ns change in the DT is up to 1 % for small ozone slant columns, increasing to ~ 4.5 % for 10 ns change in the DT.

3.2.2 Effect on TOC from direct sun measurements

The effect of DT on TOC derived from direct-sun measurements during 20 days in June 2013 at Thessaloniki (B005) and Izaña (B185) has been investigated, by applying different offsets to the nominal value of the DT that is used to correct the measured irradiances. For this analysis, the ETC has been kept constant, irrespective of the used DT. The effect of different offsets in DT on the calculated TOC is presented in Fig. 7 as function of the ozone slant column recorded at each station.

For all cases, the changes in TOC become greatest just before a new ND filter of higher optical density is set. At this point the intensity rises and the DT effect on the measured signal increases. This indicates that the effect on the calculated TOC becomes stronger for higher intensities of the direct solar irradiance. Additionally, the changes in TOC increase as the ozone slant column decreases, due to stronger intensities of incoming radiation. In accordance with the results of the sensitivity analysis presented in Sect. 3.2.1, for small changes in the DT (±2 ns) the effect on TOC derived from B185 is small, generally, below 0.5 %, and for B005 up to ~ 1.5 %. For larger changes in the DT (±10 ns) the effect on TOC is no more negligible for B185 and much stronger for B005, occasionally reaching 5 %. The stronger effect of DT on TOC derived from single-monochromator Brewers was also confirmed by Redondas et al. (2011) and Rodriguez et al. (2014).

As already mentioned, the different effects of changes in the DT on TOC measurements (as well as on the determination of the ETC) between single- and double-

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monochromator Brewers is mainly caused by the different shape of their spectral response. The spectral response determines the balance of the radiation levels at the four slits, which, in turn, affects calculated DT.

In old versions of the Brewer algorithm, there is an additional issue related to the effect of the DT on the measured signals when a ND filter is set. Before starting a direct-sun measurement, an automatic intensity check takes place in order to determine which ND filter should be used for the safe operation of the PMT. For this intensity check the signal at 320.1 nm (position 6) is measured. For double-monochromator Brewers, the signal at positions 3–5 is significantly lower than at 320.1 nm (position 6), due to the shape of the spectral response. Thus, only the signal at position 6 is actually affected by the DT when it reaches the threshold for setting a denser filter. This is not the case for single-monochromator Brewers, which have different spectral responses. For small ozone slant columns the signal at positions 4 and 5 (313.5 and 316.8 nm) is higher than at 320.1 nm and occasionally higher than the threshold. In such cases, the signals at these positions are greatly affected by the DT. As discussed in Sect. 3.1, errors in DT may induce important errors in the correction of high–intensity signals, and consequently to the derived TOC values. As the ozone slant column increases the intensity at 316.8 nm and at 313.5 nm decreases faster and gradually becomes lower than at 320.1 nm. Although the specific problem is solved in the more recent versions of the Brewer algorithm, it remains important for past datasets or for cases when an old version of the algorithm is still in use.

3.2.3 Combined effect of DT on ETC and TOC

In order to investigate the combined effect of DT errors on both the ETC derived by the Langley method and the TOC measurements, an analysis similar with the previous section is followed. Specifically, the DT effects on the ETCs that were estimated for B005 and B185 in Sect. 3.2.1 were applied to the ETC that is used in reprocessing the direct sun measurements.

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From Figs. 6–8 it appears that the errors in the calculation of the ETC lead to changes in TOC of opposite sign compared to changes on TOC due to DT errors. The effect of DT errors on the ETC is dominant for large ozone slant columns, while the effect on the calculation of TOC is dominant for small ozone slant columns. Specifically, for large ozone slant columns the results are similar with those of Fig. 6, while for small ozone slant columns the large changes shown in Fig. 7 are suppressed since the two effects are balanced. Even in this case, a 10 ns change in DT leads to 3 % change in the calculated TOC for B005.

3.2.4 Transfer of the ETC calibration from a reference instrument

The ETC is usually transferred from a reference to the calibrated instrument. To achieve that, the two collocated instruments collect a series of simultaneous TOC measurements. Possible DT errors in either the calibrated or the reference instruments may affect the calculation of the ETC. Even if the reference instrument is a well maintained and calibrated MKIII Brewer for which the DT error is negligible, it is difficult to quantify the effects of DT errors on the ETC solely from the calibrated instrument. There are two different methods for transferring the ETC from the reference to the calibrated instrument (Redondas and Rodriguez, 2012): (1) the one-point calibration, where only the ETC for the calibrated instrument is calculated and (2) the two-point calibration, where the differential ozone absorption coefficient is calculated at the same time with the ETC coefficient (Kerr et al., 1981). The effect of possible DT errors depends on the method.

As shown in Fig. 7, the DT errors affect the TOC measurements significantly when the intensity of the signal is high and/or the ozone slant column is low. The difference in TOC due to the use of an incorrect DT value cannot be eliminated simply by changing the ETC that has been derived from the incorrect DT. As it appears from Figs. 7 and 8, although a change in the ETC may partially or fully counteract the TOC errors for low ozone slant columns and high intensities, it leads to higher deviations from the reference TOC for high ozone slant columns and/or low intensities. If the two-point calibration is used the differences observed in Fig. 7 can be balanced by a combined

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change of the ETC and the differential absorption coefficient used for the calculations. This way, the change of the ETC would suppress the effect of the DT error for low ozone slant columns, while the change of the differential absorption coefficient would counteract the differences in TOC for lower slant paths due to the change of the ETC. It is obvious that the use of an incorrect DT leads to different ETCs between the one-point and the two-point calibrations. The DT effect when transferring the ETC from a reference instrument cannot be easily quantified for none of the two methods. However it is not expected to be more important than the impact of the same DT error on the ETC calculation with the Langley method.

3.3 Aerosol optical depth

Estimates of the AOD can be also derived from Brewer spectrophotometers using direct-sun measurements (Meleti and Cappellani, 2000; Kazadzis et al., 2007). We estimated that changes in the measured irradiance of 1 and 5 % lead to changes in the derived AOD of about 0.01 and 0.05 respectively for air mass close to unity. As can be seen from Fig. 4, errors of this level can be induced for very high intensities by changes in the DT of 2 and 10 ns respectively. The estimated errors in AOD are inversely proportional to air mass. Considering that the overall uncertainties in the calculation of AOD range between 0.05 and 0.07 (Kazadzis et al., 2007), only the effect of large DT errors of the order of 10 ns is important, even if the AOD has been derived for small air mass using high intensity measurements.

4 Evaluation of the dead time for past datasets: methods and difficulties

As already mentioned, in a Brewer's history there might be periods when the calculated DT may differ from the nominal by more than 2 ns. In most of these cases, repairing problems in the electronics, resetting the high voltage, or re-aligning the optics could result in suppression of the differences between the calculated DT and used DT con-

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stant (Kimlin et al., 2005). However, during regular operation it is not always easy to decide whether the derived DT is real and if its application would improve the quality of the measurements. In addition, unusual day-to-day variations of the calculated DT or indications of temperature or intensity dependence complicate further this decision. For such cases, comparison with TOC from co-located instruments or from satellites can reveal whether the Brewer measurements are properly corrected for non-linearity or not. Spectral UV irradiance is more sensitive to changes of the atmospheric parameters and the SZA; thus it is more difficult to get the same information by comparing datasets of spectral UV irradiance.

For example, in June of 2007 the preamplifier board of the PMT of B005 was replaced. Before the replacement the mean measured DT was ~ 23 ns while the DT constant was 34 ns. After the replacement the measured DT coincided with the DT constant. In order to assess whether the mean measured DT for the two different periods provides the optimal signal correction, the TOC was recalculated from intensities, then corrected with the mean measured DT and finally compared with satellite data. The comparison was made for two periods: one month before and one month after the change of the preamplifier board. Data from the NASA EOS-Aura satellite, which carries the Ozone Monitoring Instrument (OMI) were used. The specific satellite passes over Thessaloniki, where B005 is located, daily close to local noon. For the comparison only clear-sky measurements of TOC for air mass below 1.15 were used. The ratio of the TOC derived from the Brewer for DT equal to 23 and 34 ns and the OMI-TO3 (Bhartia et al., 2002) is plotted as a function of the measured intensity (from B005) at 320.1 nm. If the Brewer measurements are properly corrected for non-linearity the ratio should be independent of intensity. The ratios for the two periods, normalized with the mean over each period to remove absolute biases, are shown in Fig. 9.

For the first period, the ratio derived for the DT constant shows a clear dependence on intensity (Fig. 9c), which is practically removed when the measured DT is used (Fig. 9a). For the second period, the DT constant (which now coincides with the mea-

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sured DT) results in very small dependence from the intensity (Fig. 9d), whereas if the mean DT of the first period is used the ratio depends strongly on intensity (Fig. 9b).

In a similar study the DT for Brewer with serial number 070 was found ~ 10 ns below the DT constant, but when it was applied to the data the agreement with the TOC of the reference B183 became worse (Rodriguez et al., 2014). It must be clarified in this point that if the ratio between the ground based and the satellite TOC (or between two different ground based instruments) is independent of the intensity of the signal, this does not necessarily mean that the used DT is the real DT of the PMT. The DT correction may also compensate for instrumental malfunctions or settings that lead to real or artificial non-linear behaviour of the instrument. For example, the combination of errors in the ETC and the differential absorption coefficient might be "translated" in a DT that is different from the calculated, when the TOC from a Brewer is compared with the TOC from a satellite or another ground-based instrument. Thus the comparison with the TOC from other instruments provides an indication whether there are any remaining non-linearity issues after the correction for the DT. Since non-linearity in the measurements of TOC may not be exclusively due to DT, the DT that provides the optimal correction is not necessarily the real DT of the PMT. It is safer to check the DT with ND filters, as described in Sect. 2.3.

5 Conclusions

In this study, we assess the effects of DT on different products that are delivered by Brewer spectrophotometers, such as, spectral UV irradiance, TOC and AOD. Moreover we assess the effectiveness of different methodologies to determine accurately the DT and the applied corrections to the measured radiation signals. The analysis of data from closure experiments and long term measurements provides reliable estimates of the uncertainties associated with corrections applied for the DT effects and reveals the importance for accurate determination of the DT.

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For the theoretical point of view, the application of either the extended or the non-extended approaches on Brewer measurements provide similar estimates of the DT. However, differences are revealed when the two approaches are applied for the correction of the signal. For signals with count rates higher than $\sim 2.5 \times 10^6$ counts s^{-1} the non-extended approach results in more than 1 % lower signals compared to those derived from the extended approach for the same value of the DT. As the count rate is decreasing these differences are gradually eliminated.

In the current Brewer algorithm, nine iterations of Eq. (12) are performed for the correction of the measured signals. Here we have found that the corrected signal converges already after 5 iterations, independently of intensity and the value of DT, for both the extended and the non-extended approaches.

The correction of the dark signal for the DT effect was found unnecessary, as long as the level of the dark signal remains below 10^4 counts s^{-1}.

Further evaluation of the current algorithm for the determination of the DT indicates that the results are accurate as long as the signal at slits 3 and 5 is of similar level (i.e. within a factor of 2). If the ratio between the count rates is outside these limits, then the DT constant is underestimated. Increasing the signal and the number of cycles reduces the noise and the uncertainty of the final products. Specifically, as long as the signal level remains above 10^6 counts s^{-1}, measurements with 10 cycles are sufficient to keep the uncertainty of the calculated DT below 3 % ($\sim$ 1 ns).

Measurements of the direct solar irradiance (which is usually stronger than the radiation of the Brewer's internal standard lamp) provide more accurate estimates of the DT with lower uncertainty. In order to achieve that, the measurements should be performed at wavelengths with N3/N7 ranging between 0.3 and 0.7, and signal level above 10^6 counts s^{-1}. Since at some locations or seasons direct sun measurements might not be achievable for long time, these should be used only complementarily to the standard lamp-based determination of the DT. Occasionally, the standard lamp-based DT can be very noisy so that the derived values might not be the most suitable for correction of the signals. In such cases, and if direct sun-based estimates of the DT are not available,

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the DT can be determined by optimizing the spectral transmittance of ND filters derived from measurements that are corrected with different DT values.

An independent check of the DT that provides the optimum correction to the measured signals was achieved from short-term comparisons of the derived TOC with data from satellites or from co-located stable instruments. However, this method might not be very safe. Errors in parameters that are used to derive TOC (e.g. ETC, differential absorption coefficient) might lead to artificial non-linearity in the final products which is subsequently balanced by the use of an incorrect DT when the data are compared against data from reference instruments.

Considering the correction of the UV irradiance, even an error of 10 ns in the DT does not induce errors greater than 1.5 % on the calculated irradiance, as long as the count rate remains below 10^6 counts s^{-1}. The maximum count rate for global or direct-sun UV scans does not usually exceed 3–3.5 × 10^6 counts s^{-1}. As the incident irradiance is getting closer to this limit the errors are becoming more important. For such high count rates, a 2 ns error in the DT results into an error of ∼ 1 % in the irradiance which rises to ∼ 5 % for 10 ns error in the DT. For the calculation of TOC, the uncertainties related to the DT are highly dependent on the shape of the instrument's spectral response; thus on the type of the instrument. For the double-monochromator Brewers, the error in TOC does not exceed 2 %, even for 10 ns error in the DT, while for single-monochromator Brewers the error may escalate to ∼ 5 %. The tolerance of 2 ns suggested by the manufacturer for the DT error (Kipp & Zonen Inc., 2008; SCI-TEC Instruments Inc., 1999) has negligible impact on TOC for double-monochromator Brewers, and up to 1 % for single-monochromator Brewers. As the target for the total uncertainty in TOC measurements is 1 % (Kerr et al., 1985), it is obvious that this tolerance for the DT has to be lowered. The effect of DT errors in the calculation of AOD is found to be of less importance compared to errors in UV irradiance and TOC.

Based on the results of this study we can summarize the following suggestions:

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For the calculation of the dead time from measurements of the sun, these should be performed for wavelengths and SZAs that ensure comparable signals at the two slits (3 and 5), and for signals at slit 7 that remain above 10^6 counts s^{-1}.

For the correction of the signal the nine iterations of Eq. (12) can be reduced to five without affecting the quality of the Brewer products. This will reduce the time required for several operational routines.

Regarding the TOC measurements from single-monochromator Brewers, the tolerance of 2 ns in the DT error should be changed to 1 ns. Additionally, before a ND filter is set, the intensity for both the 320.1 and the 316.8 nm should be checked in order to keep the maximum signal at all slits below the defined threshold.

Lowering the intensity threshold for both the single- and the double-monochromator Brewers would lead to smaller uncertainties in TOC and AOD due to DT errors. During global spectral irradiance measurements the signal may reach levels ($\sim 3.5 \times 10^6$ counts s^{-1}) where the impact of the DT errors is becoming very important. However, using a ND filter in order to limit the impact of the DT errors might result to increased uncertainties due to errors in the determination of the filter transmittance (Redondas et al., 2011).

This study has been accomplished in the framework of COST 1207 which aims to establish a coherent network of European Brewer Spectrophotometer monitoring stations and among others to harmonize operations and achieve consistency in quality control and quality assurance. The results and the suggestions of the present study will hopefully contribute to improve the quality of the Brewer products. Given that some Brewers are in operation since the early 1980's, more accurate DT correction would lead to more accurate detection of trends in ozone, global UV irradiance and other products, and to more reliable data that can be used for the validation of satellite products, and for several applications in physical (Erickson III et al., 2015) and health (Lucas et al., 2015) sciences.

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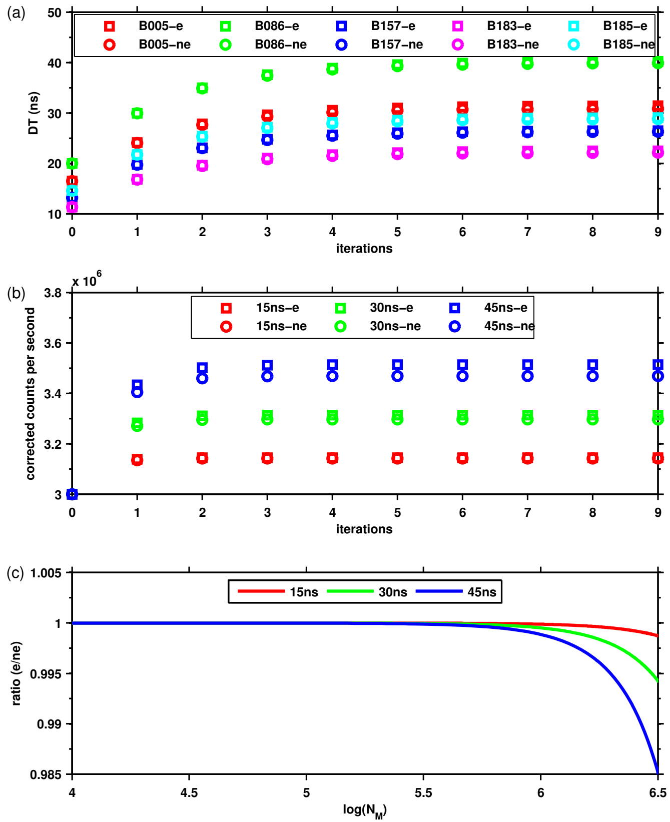

Figure 1. (a) Extended (e) and non-extended (ne) DT for five Brewers with DT constants ranging from 19 to 42 ns as a function of the number of iterations. **(b)** Corrected counts/sec for different number of iterations and different DT. **(c)** Ratio between the extended and the non-extended DT as a function of the logarithm of the measured, uncorrected for the DT, count rate.

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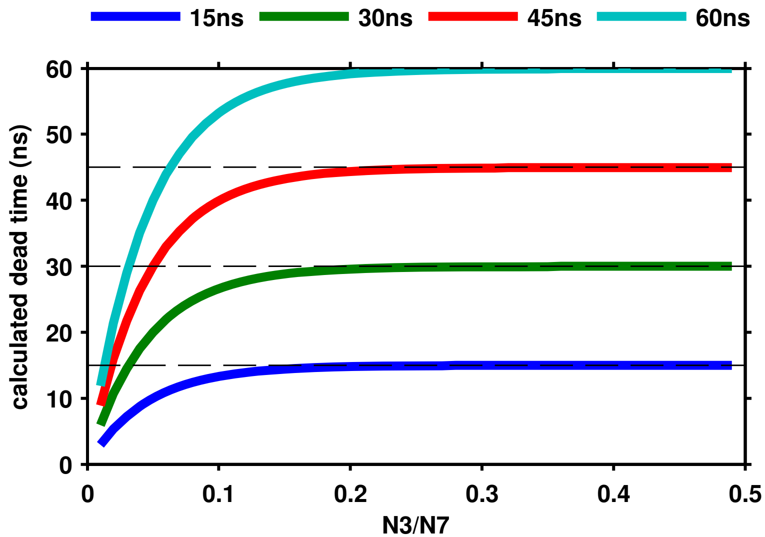

Figure 2. DT as it is calculated with the standard Brewer algorithm for different N3/N7 ratios and for 4 different reference DT values. The results are for photon rate (at position 7) of 10^6 counts s^{-1}.

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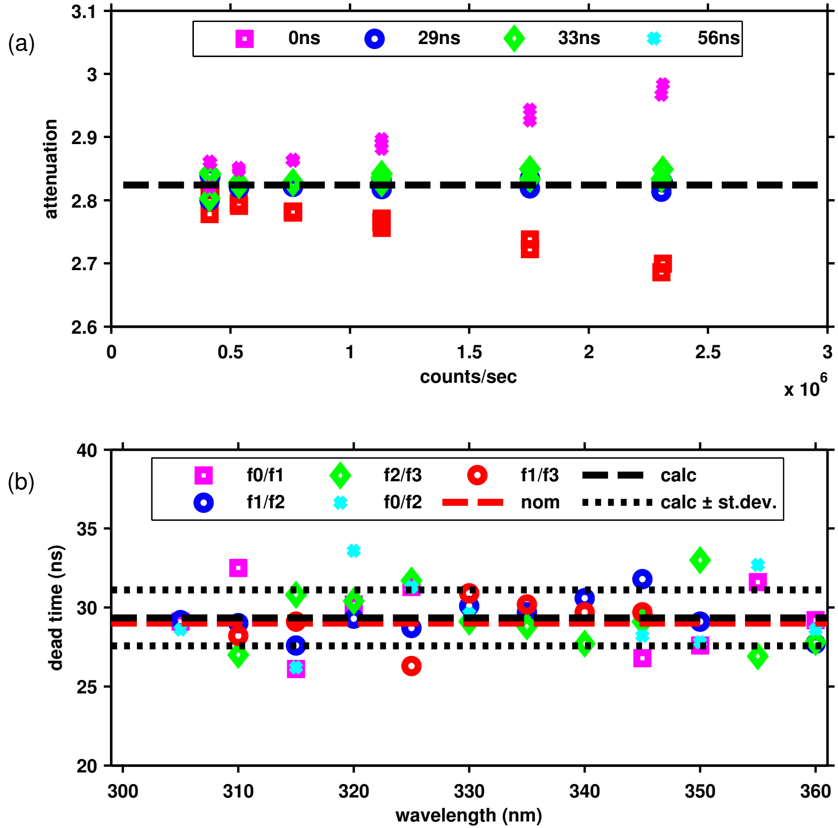

Figure 3. (a) Relative attenuation between positions 0 and 1 of FW#2 for irradiance signals at 360 nm. **(b)** The optimum calculated DT value for different wavelengths and different relative attenuations (pairs of ND filters). The derived mean values and the used DT constant are also shown. All the results are for B185.

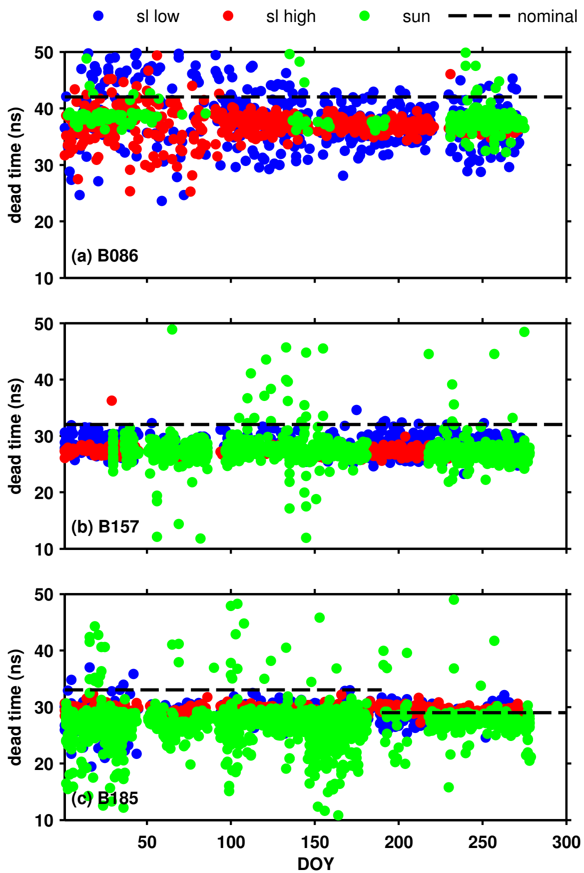

Figure 4. DT calculated from measurements of the standard lamp (high and low intensity) and the sun for three Brewers **(a)** 086, **(b)** 157, and **(c)** 185. The results are for 2014 and are presented as a function of DOY. Dashed lines represent the DT constant used by the instruments in each period.

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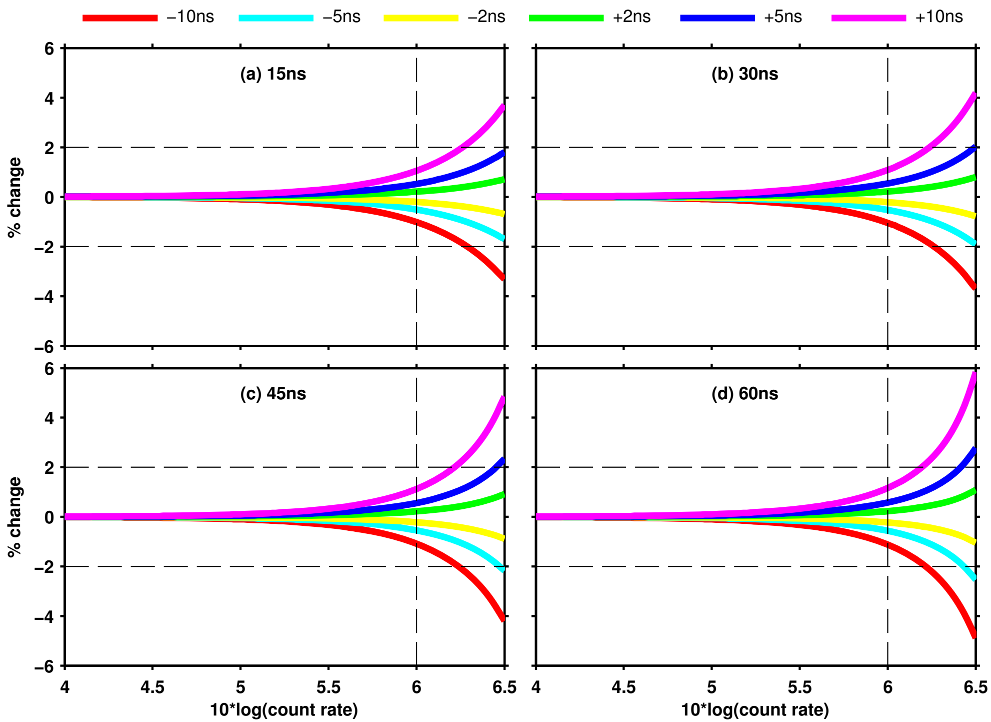

Figure 5. Changes in UV irradiance as a function of intensity due to errors in the determination of the DT, for different values of the reference DT: **(a)** 15 ns, **(b)** 30 ns, **(c)** 45 ns, and **(d)** 60 ns.

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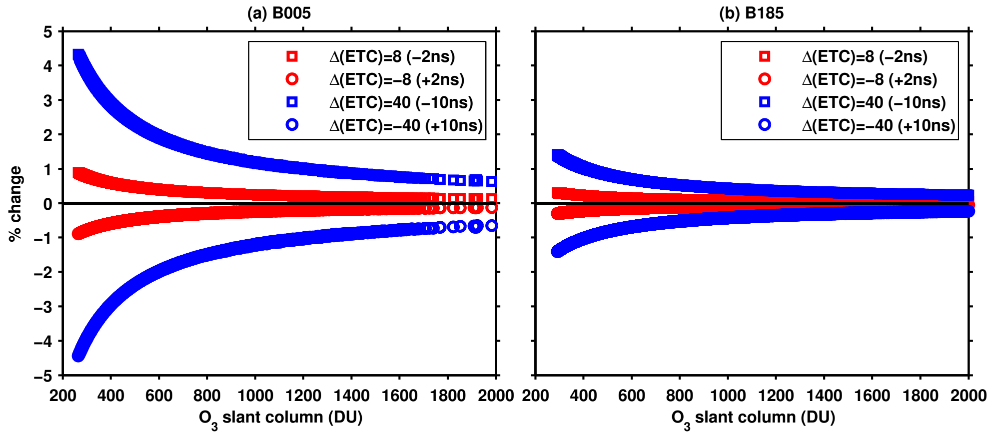

Figure 6. Changes (%) in the calculated TOC as a consequence of the ETC change due to ±2 and ±10 ns change in the DT, as a function of slant column of ozone. Results are presented for B005 **(a)** and B185 **(b)**. For B005 the used ETC has been changed by ±8 units for a ±2 ns change of the DT and by ±40 units for a ±10 ns change of the DT. For B185, the corresponding changes of the ETC are ±3 and ±15 units.

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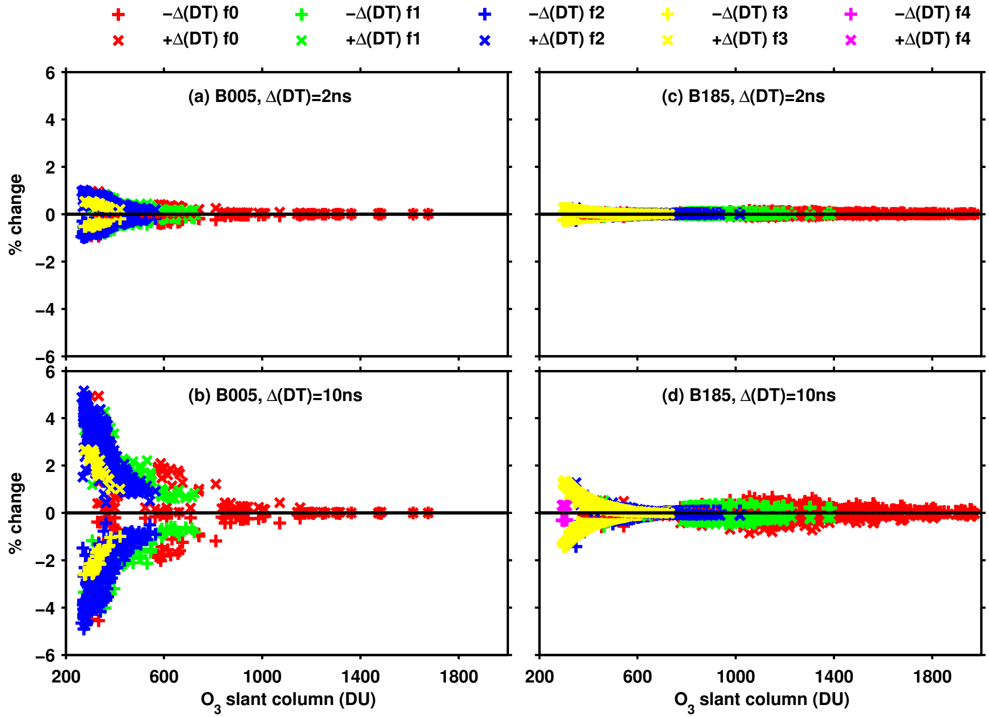

Figure 7. Changes (%) in TOC calculated from direct-sun measurements due to ±2 ns **(a, c)** and ±10 ns **(b, d)** changes in the DT, as a function of ozone slant column, for B005 **(a, b)** and B185 **(c, d)**. Different colors refer to data measured without (red) and with neutral density filters of optical density 0.5 (green), 1 (blue), 1.5 (yellow) and 2 (magenta). The reference DT is 34 ns for B005 and 29 ns for B185.

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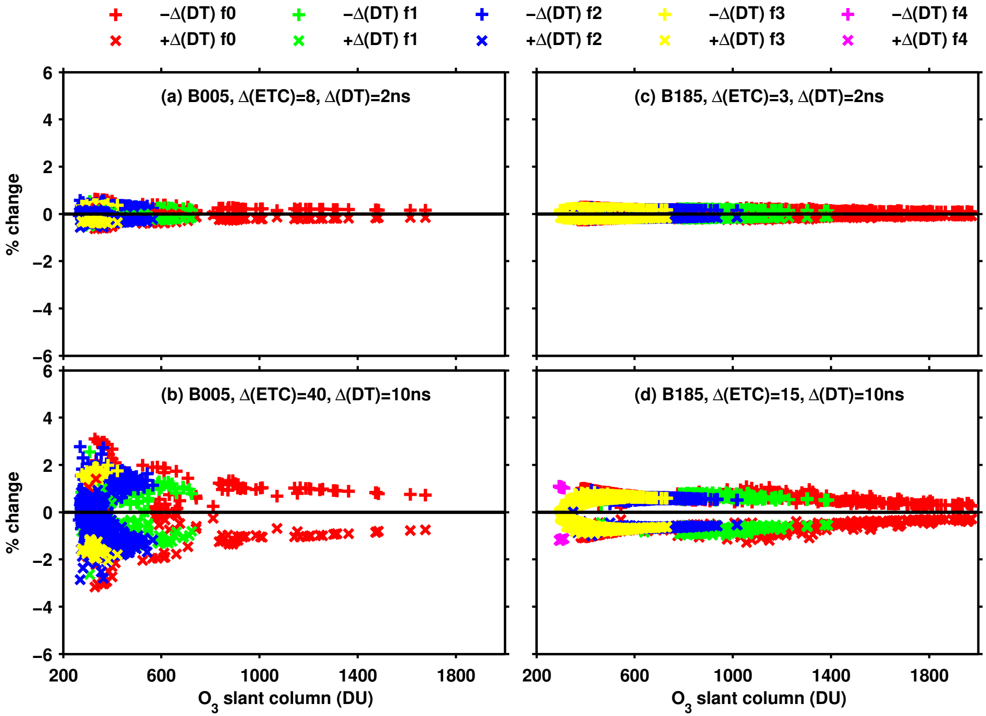

Figure 8. Changes (%) in TOC calculated from direct-sun measurements due to ±2 ns **(a, c)** and ±10 ns **(b, d)** changes in the DT, as a function of ozone slant column, for B005 **(a, b)** and B185 **(c, d)**. For each change of the DT, the ETC used for the calculations is subjected to the changes described in Sect. 3.2.1. Different colors refer to data measured without (red) and with neutral density filters of optical density 0.5 (green), 1 (blue), 1.5 (yellow) and 2 (magenta). The reference DT is 34 ns for B005 and 29 ns for B185.

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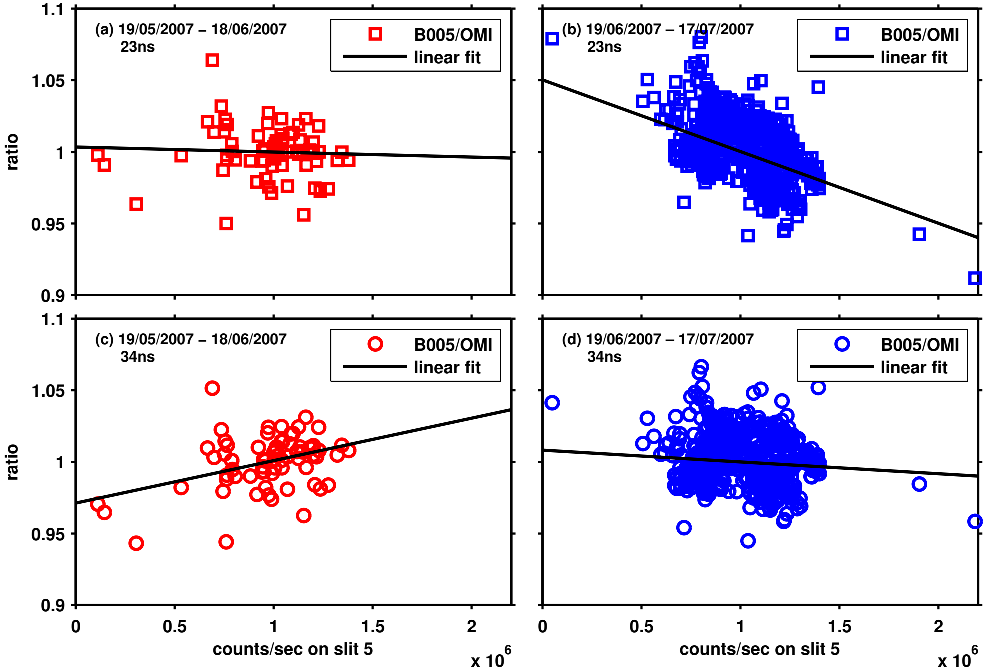

Figure 9. Ratio of TOC derived from B005 for different DT values and OMI, as a function of the Brewer intensity at 320.1 nm.

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