Wavelength calibration of Brewer spectrophotometer using a [..*] <u>tuneable</u> pulsed laser and implications to the Brewer ozone retrieval

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Abstract.

In this contribution we present the wavelength calibration of the traveling reference Brewer spectrometer of the Regional Brewer Calibration Center [$..^2$](RBCC-E) at PTB in Braunschweig, Germany. The wavelength calibration is needed for the calculation of the ozone absorption coefficients used by the Brewer ozone algorithm. In order to validate the standard procedure

- 5 for determining Brewer's wavelength scale, a calibration has been performed by using a [..³]tuneable laser source at PTB in the framework of the EMRP project ENV59 ATMOZ "Traceability for the total column ozone". Here we compare these results to those of the standard procedure for the wavelength calibration of the Brewer instrument. Such a comparison allows validating the standard methodology used for measuring the ozone absorption coefficient with respect to several assumptions. The results of the laser-based calibrations reproduces those obtained by the standard operational methodology and shows that there is [..⁴
- 10]a underestimation of 0.8% [..⁵]due the use of the parametrized slit functions.

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1 Background

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[..⁶] The wavelength calibration is needed for calculation of the ozone absorption coefficient used by the Brewer ozone retrieval algorithm. The Brewer spectrophotometer has two operating modes. In the ozone mode, used for the total ozone column measurements, [..⁷] the diffraction grating stays at a fixed position while the six operational wavelengths are selected by a rotating slit mask[..⁸]. The scanning mode is used to perform spectral irradiance measurements in the [..⁹] ultraviolet (UV)

5 spectral range. In this mode, the slits are fixed and the spectral scan is carried out by turning the diffraction grating. The usual wavelength calibration procedure is performed in the scanning mode by analyzing recorded emission lines of the spectral discharge lamps, which are usually mercury (Hg), cadmium (Cd), and zinc (Zn). [..¹⁰]Using the spectral lines provided in Table 1 allow us to determine the central wavelengths and the corresponding FWHM (full width at half maximum[..¹¹]) of the slit functions as well as the relation between the positions of the grating and the corresponding instrument wavelengths

10 (dispersion relation) required to determine the operational wavelengths used for the ozone determination. To obtain the ozone absorption coefficients, the instrumental slit functions are convolved [..¹²] whit the Bass & Paur ozone absorption cross-section data. The use of [..¹³] the of the tuneable laser source allow us to:

Calculate the ozone absorption coefficients from the calibration directly in the ozone mode. The normal determination of
the ozone absorption coefficients involves scanning of the spectral lines in the scanning mode of the instrument so that

- the dispersion relation is required to convert the grating positions in micrometer steps to the respective wavelengths. Here we can determine the instrumental slit functions directly in the ozone mode by scanning them with the [..¹⁴]tuneable laser and weight them with the ozone absorption cross-sections without the need [..¹⁵]for the assumptions about the slit functions and the dispersion relations used in the normal calibration procedure[..¹⁶]
- 20

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Calculate the dispersion relation based on regularly spaced reference spectral lines provided by the [..¹⁷]tuneable laser instead of the irregularly distributed emission lines of the Hg, Cd and Zn spectral lamps.

During the experiment we performed three measurements:

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⁶removed: Nowadays the primary ground-based instruments used to report total ozone column (TOC) are Dobson and Brewer spectrophotometers. Based on the irradiances measured by these instruments in the ultraviolet (UV) spectral range and on well-defined retrieval procedures, TOC values are derived. The Brewer spectrometer (Brewer, 1973; Kerr et al., 1981; Kerr, 2010) was introduced in the 1980s as an automatic device measuring direct solar UV radiation and global UV irradiance. Both the Brewer and the Dobson instruments were considered by the World Meteorological Organization (WMO) in the framework of the Global Atmosphere Watch program (GAW) as the standard instruments for TOC monitoring. The

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Table 1. Emission lines of the discharge lamps used for Brewer calibration

Lamp	Line (nm)	Slits
Mercury (Hg)	289.36	0–1
Hg	296.728	0–3
Zinc (Zn)	301.836	0–5
Zn	303.578	0–5
Cd (multiplet)	308.082	0-5
Cd	313.3167	0–5
Cd	326.1055	0–5
Zn	328.233	0–5
Hg	334.148	0–5
Cd	340.3652	0–5
Cd	349.995	4–5
Cd (multiplet)	361.163	5

1. The standard method for the dispersion measurements using spectral lamps described in section 2.

2. Direct dispersion measurements (laser scanning).

Direct dispersion measurements (laser scanning). While the Brewer [$..^{18}$] is measuring in the ozone mode and in the aerosol mode, the laser wavelength is swept ± 2 nm with a step of 0.04nm around the six Brewer operational wavelengths selected by the rotating slit mask for different grating positions (Figure 1)

3. Dispersion measurements using the tunable laser (Brewer scanning). While the laser is emitting at a fixed wavelength[..¹⁹][..²⁰], the Brewer [..²¹]scans ±2nm around [..²²] this wavelength in the scanning mode by moving the grating and using the 6 slits. Such Brewer scans are carried out at wavelengths ranging from 290 nm to 365 nm with an increment of 5nm. The results allow us to estimate the dispersion approximation error due to the lack of spectral lines [..²³] at the end of spectral range of the Brewer spectrophotometer and due to the fact that the emission lines of the used lamps are not equally spaced.

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2 Calibration of the Brewer spectrophotometer

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The Brewer instrument measures the irradiance of direct sunlight at six nominal wavelengths (λ) in the UV range (303.2, 306.3, 310.1, 313.5, 316.8, and 320.1[..⁵⁶] nm), each spectral band covering a bandwidth of 0.5 nm ([..⁵⁷] resolution power $\lambda/\delta\lambda$ of around 600). The spectral resolution is achieved by a holographic grating in combination with a slit mask that selects the channel to be analyzed by a photomultiplier tube (PMT). The longest four wavelengths are used for the ozone calculation.

Based on the Lambert-Beer law, the total ozone column in the Brewer algorithm can be expressed as:

$$X = \frac{F - ETC}{\alpha \mu} \tag{1}$$

where F are the measured double ratios corrected for Rayleigh effects, α is the ozone absorption coefficient, μ is the ozone air mass factor, and *ETC* is the extra-terrestrial constant. The F, α and *ETC* parameters are weighted functions at the operational wavelengths with weighting coefficients [...⁵⁸]w:

$$F = \sum_{i}^{4} w_i F_i - \frac{p}{p_0} \beta_i \mu \tag{2}$$

$$\alpha = \sum_{i}^{4} w_i \alpha_i \tag{3}$$

$$ETC = \sum_{i}^{4} w_i F_{0i} \tag{4}$$

where, β_i are the Rayleigh coefficients, p is the climatological pressure at the measurement site, p_0 is the pressure at sea 15 level, and F_0 are the individual extra-terrestrial constants at each wavelength. The weights w = [1, -0.5, -2.2, 1.7] have been chosen so as to minimize the influence of SO₂ and verify:

$$\sum_{i}^{4} w_i = 0 \tag{5}$$

$$\sum_{i}^{4} w_i \lambda_i = 0 \tag{6}$$

This widely eliminates absorption features which depend, in local approximation, linearly on the wavelength, like for exam-20 ple the contribution from aerosols.

We can divide the calibration in three steps including instrumental calibration, wavelength calibration, and ETC transfer:

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- 1. The instrumental calibration includes all the parameters that affect the $[..^{59}]$ measured signal counts $(F)[..^{60}]$, in particular, photomultiplier dead time correction, temperature coefficients and filter attenuation.
- 2. Wavelength calibration is needed to determine the ozone absorption coefficient. The so-called "dispersion test" is used to obtain the particular wavelengths for the instrument and the slits, or instrumental functions, of each spectrophotometer. Note that the precise wavelengths of every Brewer spectrophotometer are slightly different from instrument to instrument.
- 5 Note that the precise wavelengths of every Brewer spectrophotometer are slightly different from instrument to instrument.
 - 3. Finally, the ETC transfer is performed by comparison with the reference Brewer instrument or, in the case of the reference instruments, by the Langley method.

The Brewer wavelength calibration follows the operative procedure (Gröbner et al., 1998; Kerr, 2002) used by the Regional Brewer Calibration Center-Europe (RBCC-E) at the calibration campaigns. In summary, the individual wavelengths (bands) in the Brewer instrument are selected through the use of a stainless steel mask of seven slits located at the focal plane of the

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in the Brewer instrument are selected through the use of a stainless steel mask of seven slits located at the focal plane of the spectrometer. The particular wavelength is determined by analyzing the measurements of a series of discharge lamps during so-caled dispersion test, which determines the central wavelength and FWHM of every slit. Then the wavelength setting is optimized to minimize the effect of wavelength shift during the operation of the instrument by performing the so-called sunscan test (Lamb, K and Asbridge, A.I., 1996). Finally, the ozone absorption coefficient is determined for every slit.

15 The ozone absorption coefficient were defined as (Vanier and Wardle, 1969; Bernhard et al., 2005; Wardle, 2008):

$$\widetilde{\alpha}(X,\mu) = \sum w_i[..^{61}] \frac{\int \alpha(\lambda) * \mathsf{S}(\lambda,\lambda') * \mathsf{F}(\lambda,\lambda',\mathsf{X},\mu) \mathsf{d}\lambda'}{\int \mathsf{S}(\lambda,\lambda') * \mathsf{F}(\lambda,\lambda',\mathsf{X},\mu) \mathsf{d}\lambda'}$$
(7)

Where S is the instrumental slit function for the corresponding wavelength, [..⁶²] F is the sun spectrum [..⁶³] that depends mostly on the ozone concentration and airmass, and σ is the ozone cross-section at the temperature of -46.3 °C for Dobson and at -45 °C for Brewer instruments.

- 20 The Brewer operative method uses the following assumptions:
 - 1. Use "ideal" slits; the slit functions are parametrized as trapezoids, i.e., isosceles triangles truncated at 0.87 height.
 - 2. Stray light is not considered, i.e., zero slit function values are assumed outside the triangle.
 - 3. The FWHM of the triangle is dependent of the slit and it is derived from the dispersion test.
 - 4. The ozone cross sections are expressed by the Bass & Paur absorption coefficient data set.

25 5. Solar spectrum is not considered, $([..^{64}]F==1)$

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Figure 1. $[..^{66}]$ Brewer measurements in the ozone mode $[..^{67}]$ while the laser wavelength $[..^{68}]$ is changed every $[..^{69}]$ 0.1 nm. The central wavelength and the FWHM calculated are displayed in red using the same methodology of the dispersion test $[..^{70}]$

Under these assumptions, the ozone effective absorption is essentially obtained the same way as in the approximation method of Bernhard et al. (2005) used with Dobson spectrophotometers. (see Eq. 8).

$$\alpha_{i} = [..^{65}] \frac{\int \sigma(\lambda) S_{i}(\lambda) d\lambda}{\int S_{i}(\lambda) d\lambda}$$
(8)

2.1 Dispersion Test

5 The Brewer spectrophotometer is constructed based either on a single or a double [..⁷¹]monochromatoror of modified Ebert–Fastie type, generally [..⁷²]referered to as single or double Brewer, respectively. The first monochromator disperses the incoming radiation onto six exit slits. In the case of the [..⁷³]double Brewer, the six exit slits (intermediate slits) of the first monochromator are the entrance slits to a second monochromator that is used in subtractive mode. The wavelength is selected by choosing one of the six exit [..⁷⁴]slit (ozone mode) or rotating the grating (scanning mode). The rotation of the grating is managed by a drive mechanism consisting of a motor-driven micrometer linked to an arm that rotates the grating. The smallest

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wavelength increment corresponding to one stepper motor step varies steadily from approximately 8.0 pm to 7.0 pm [$..^{75}$](0.0080 nm) Gröbner et al. (1998).

[..76]

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The dispersion relation, which provides the relation between the micrometer steps and the monochromator set wavelengths, is determined by scanning the emission lines as described in section 1. The line scans are carried out with with an increment of 10 motor steps (0.7[..⁷⁷]A). From the results, the central position and the FWHM of the slit function are calculated in motor steps assuming an isosceles triangle. The both sides of the peak are fitted to a straight line taking only the function values above 20% and bellow 80% of the normalized peak. The central point is calculated by the intersection point and the FWHM is the

width of the triangle (Figure [..⁷⁸]??). Finally, the dispersion relation is calculated using a quadratic polynomial or the [..⁷⁹

10]spline cubic approximation. This relation is used to transform the previously determined central positions and FWHMs of the slit functions in micrometer steps to a wavelength scale.

 $[..^{80}]$

The cubic method of Gröbner et al. [..⁸¹] use the optical design of the [..⁸²] brewer to transfer line measures from one slit to the other slits [..⁸³] reducing the number of free parameters to [..⁸⁴] adjust compared with the quadratic method. However

15 [..⁸⁵] there is a systematic difference [..⁸⁶] on the calculation of the ozone absorption coefficient between the two methods.
 Both methods generally agree [..⁸⁷] if only the brewer ozone spectral range [..⁸⁸] is used (Redondas and Rodriguez-Franco, 2012).

The stability of the wavelength calibration during Brewer operations is checked by measuring the internal Hg lamp. In most of the Brewers, the 302 nm double line (302.150 nm and 302.347 nm) is used due to its proximity to the Brewer operational wavelengths. However, for Brewer #185 and for an increasing number of other Brewers the test is performed using the more powerfull 296.7 nm line. The wavelength test includes 12 measurements of the line from the mercury lamp on slit 0, with an

⁷⁵removed: (Gröbner et al., 1998)

⁷⁶removed: Slit function of the slit #3 (320 nm) measured in the scanning mode: the standard method uses the normalized values only between 0.2 and 0.8 (points with crosses). The figure shows the signals recorded during the scans (blue diamonds) and the non-linearity-corrected values (black circles) in counts/second. The hysteresis is evident from the asymmetry of the uncorrected signals in the low signal region of the plot. The center steps and the FWHMs calculated from the up-scan are shown in red while the values derived from the down-scanned slit function are presented in blue text. The central wavelength determination is not affected by the nonlinearity but the apparent FWHMs would be larger if the non-linearity correction were not applied. The resulting parametrized slits are represented on the right axis.

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Figure 2. Measured slit function of the 320 nm (slit #3): the method uses the normalised values only between 0.2 and 0.8 (point with cross). The calculated center step and the FWHM are shown for the up (red) and down scan (blue). The resulting parametrized slit are represented on the right axis. Uncorrected counts/second (blue diamonts) and non-linearly-corrected values (black circles). The central wavelength determination is not affected by the nonlinearity but the FWHM is bigger if the correction is not applied. The hysteresis is evident from the asymmetry of the function at the low values region of the plot.

increment of ± 10 steps of the micrometer motor. The obtained curve for the line peak is compared to a stored reference one. The comparison is done by shifting the two [..⁸⁹]scaned curves against each other and calculating the correlation coefficient between the two after each shift. The interpolated step number yielding the maximum of the correlation coefficient provides the reference micrometer position (Savastiouk, 2005). If the required adjustment of the micrometer position is more than one

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and a half motor steps, the test is repeated. [..⁹⁰] The accuracy of the wavelength setting of the Brewer instrument achieved by such an approach is [..⁹¹] limited by one and half motor steps and cannot be better than about 0.86A. This affects the ozone [..⁹²] abortion coefficient by approximately of [..⁹³] 0.10 atmcm⁻¹ and the ozone concentration by 0.3%.

3 Pulsed laser-based measurements

3.1 Instrumental setup

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 $^{^{91}}$ removed: defined by a rectangular probability distribution function on the interval of +-0.5 steps. Thus, the respective standard uncertainty is 0.5/sqrt(3)= 0.289 steps. During the brewer operation at RBCC-E calibration campaigns the ozone observations are discarded if the subsequent hg test is repeated, the bigger discrepancy for the accepted measurements will be 1.5 steps. This 1.5

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Figure 3. Pulsed optical parametric oscilator (OPO)-based setup at PTB that was used for measuring the slit functions of the Brewer spectrophotometer.

[...⁹⁴]For scanning the bandpass functions of the Brewer instrument, an upgraded PLACOS setup (Nevas et al., 2009) featuring a [...⁹⁵] tuneable pulsed laser system based on an Optical Parametric Oscillator (OPO) was used at Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig (Figure 3). The new OPO system generates 3-6 ns pulses at 1 kHz repetition rate in the spectral range from 210 nm to 2600 nm. The laser wavelengths were monitored during the measurements by a wavemeter (laser spectrum analyzer) and a high-resolution spectrometer with an uncertainty of 0.01 nm. The laser beam was guided into the direct port of the Brewer spectrophotometer by using a liquid light guide. A fraction of the beam was directed to a monitor photodiode in order to account for the output power changes of the laser beam. The photocurrent of the silicon photodiode was measured by [...⁹⁶][..⁹⁷][..⁹⁸][..⁹⁹] a charge meter.

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⁹⁶removed: an electrometer. During the measurements, the laser wavelengths were monitored by a wavemeter (laser spectrum analyzer) and a high-resolution spectrometer with a FWHM of 0.1

⁹⁷removed: . The wavelength accuracy of the wavemeter according to the manufacturer's specifications is between 6 pm and 3 pm in the spectral range from 290

⁹⁹removed: , respectively. The calibration of the instrument is accomplished by means of an autocalibration procedure based on an internally built-in neon lamp. The wavelength scale of the high-resolution spectrometer was calibrated by using the spectral emission lines of a Hg lamp. The wavelength uncertainty of the calibrated spectrometer in the above-mentioned spectral range was estimated to be 10 pm. A side-by-side comparison of the laser wavelength values measured by both instrument types showed an agreement well within 10 pm.



Figure 4. Log-log plot of the normalized ratio of measured Brewer counts to the monitor photodiode signal, which is proportional to the laser power, plotted as a function of the Brewer counts/s. The black points shows the measured points while the red curve is the fit used to correct the Brewer signals for the nonlinearity.

In contrast with the standard calibration procedure, where the Brewer instrument scans the lines of the spectral lamps, in this experiment the Brewer measures in ozone mode. Here, the Brewer grating is fixed at the ozone position while the coupled laser beam is measured using the seven slits (slit #1 is used to obtain the dark signal values). During these measurements the wavelength of the OPO system is scanned with 0.04 nm step. The experiment is complemented by the measurements in the

5 Brewer scanning mode, where the [..¹⁰⁰]tuneable laser is used as a source of spectral lines covering the range from 290 nm to 360 nm on a regular grid with 5 nm step.

¹⁰⁰removed: tunable

Brewer ozone mode raw counts mic=1020



Figure 5. Brewer measurements in the ozone mode while the laser wavelength is changed every $[..^{104}]0.1$ nm. The $[..^{105}]$ blue curve corresponds to the dark counts obtained from the measurements of slit 1.

3.1.1 Non-linearity of the PMT

The Brewer detector system, which is based on a PMT, responds non-linearly to pulsed sources. For the measurements of pulsed sources, the PMT manual advises to change the electronics configuration. As the main objective was to validate the operational wavelength calibration of the Brewer, we decided to keep the instrument configuration equivalent to that during the

5 field operations. The non-linearity problem was solved by determining the respective correction function. For this purpose, the power of the laser beam was varied while simultaneously measuring signals of the PMT and the linear monitor photodiode.

The ratios of the measured Brewer counts to the recorded monitor photodiode signals are shown in figure 4. The nonlinearity is evident in the figure together with hysteresis region near 10^4 Brewer counts/[..¹⁰¹]seconds. The correction is not reliable around 10^3 counts/[..¹⁰²]seconds and lower than 100 counts/[..¹⁰³]seconds. As we can control the power of the laser

10 beam, it is possible to work on the "flat regions" of the non-linearity characteristics and apply the determined correction. This correction does not affect the calculated central wavelength of the slit functions, though, it does affect the determined FWHM values (Figure 2) if the correction is not applied.

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Table 2. Ozone absorption [..¹¹²] coefficient in atm cm^{-1} calculated using four [..¹¹³] absorption [..¹¹⁴] cross sections.[..¹¹⁵]

	[116]Parametrized	[¹¹⁷][¹¹⁸][¹¹⁹][¹²⁰]Measured
BRW	0.3381	[¹²¹][¹²²][¹²³][¹²⁴] 0.3407
B&P	[¹²⁵]0.3330	[¹²⁶][¹²⁷][¹²⁸][¹²⁹] 0.3360
DMB	0.3483	$[^{130}]0.3514[^{131}][^{132}]$
[¹³³]SDK	[¹³⁴]0.3392	[¹³⁵]0.3422[¹³⁶][¹³⁷]

[..¹⁰⁶]Fron the Figure 5 we can see that correction is not reliable for signals lower than 100 counts/seconds. Also we observed that the recorder dark signal values (measurements performed with the blocked slit #1) were highest immediately after exposing the PMT to the laser light. The dark signal of the PMT was then gradually fast decreasing with time after the excitation, which may cause the signal values obtained for slit #1 (measured immediately after slit #0) be higher than for the other slits measured afterwards.

3.1.2 Slit parametrization

The Brewer algorithms assume trapezoidal slits functions cut at 0.87 of the height (Figure 6) with the center wavelength and the FWHM calculated for every slit from the dispersion relation. The laser measurements allow us to estimate the effect on the ozone calculation if we use the directly measured slit functions instead of the [$..^{107}$]parametrised ones. For this purpose we

10 calculate the ozone absorption coefficients for the four ozone cross sections evaluated in the "ACSO" comitee ("Absorption Cross Sections of Ozone") (Orphal et al. (2016)[..¹⁰⁸].

Among the available data sets there are versions of Bass and Paur (1985) cross-sections denoted as Brewer operational (Brw), IGACO quadratic coefficient (B&P), the cross-sections of Daumont Brion Malicet (DBM) (Daumont et al. (1992), Brion et al. (1993), and Malicet et al. (1995)), and the newly recommended data set for ozone ground-based calculation by Serdyuchenco, Groshelev, Weber (SGW) (Serdyuchenko et al., 2012; Gorshelev et al., 2012; Weber et al., 2016).

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Using the measured slit functions, the calculated effective ozone cross sections are $[..^{110}] 0.9\%$ higher compared to those obtained by using the parametrized Brewer slits in the standard procedure (Table 2) $[..^{111}]$ independently of the cross sections

used.

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¹¹⁰ removed: 0.85%

¹¹¹removed: , this difference is reduced approximately to the half when a triangular parametrization is used 0.44%, the results are very similar for all the cross section with the exception of the Bass & Paur used by the brewer due his low resolution



Figure 6. Plot of the parametrized (thick lines, left axis) and the measured slit functions (dots, left axis) as well as the different ozone [..¹⁰⁹] cross sections in cm^{-1} (right axis) used for the Brewer effective ozone absorption coefficient calculation.

4 Discussion

The experiment allows us to validate the Brewer standard methodology used to perform the wavelength calibration. For this purpose, we compare laser-based wavelength calibration results to those yielded by the standard operative method based on scanning the spectral lamps in case of both the quadratic and the cubic fit to the dispersion relation.

Figure [..¹³⁸]7 shows discrepancies between the central wavelengths calculated by the quadratic and the cubic fits to be bigger than 0.1Å for wavelength above 320 nm and much bigger near 350 nm. This is also indicated by a systematic shape of the residuals of the quadratic fit that are much larger than for the cubic fit ([..¹³⁹]see figures 9 and 10). This indicates that the quadratic fit is only valid in the ozone range ([..¹⁴⁰]310-320 nm) as already noted in Gröbner et al. (1998). The comparison of the calculated FWHMs (Figure [..¹⁴¹]8) shows a different pattern with a difference of 0.1Å between the direct and the scanning methods in the ozone range and with a smaller difference between the quadratic and the cubic fits.

 $[..^{142}]$

¹³⁸removed: ??

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Figure 7. Differences between the central wavelengths determinated directly and by the scanning methods: with the laser wavelengths in equally spaced grid every 5nm and lines of the discharge lamps; in both cases quadratic and cubic fitting are used.





The differences to the directly calculated ozone absorption coefficients [$..^{143}$] are summarized for the six measurements in [$..^{144}$] table 3, taking as a reference the direct measurements. The quadratic fits result in bigger differences of around 1% whereas in the case of the cubic fits the differences decrease to 0.3% and 0.1% when the laser or the discharge lamps are used, respectively.

¹⁴³ removed: calculated from the scanning and the direct measurements of the slit functions

¹⁴⁴removed: Table

	brw_scan^1	opo_quad^2	opo_cubic^3	$lamp_quad^4$	$lamp_cubic^{E}$
SGW	0.3409	0.3442	0.342	0.3446	0.3412
ratio	1	0.9881	1.0033	1.0108	1.001

Table 3. [..¹⁴⁵]Ozone absorption coefficient in [..¹⁴⁶][..¹⁴⁷] atm cm^{-1} calculated using four absorption cross sections

1. Laser wavelength scanned @ fixed Brewer grating position

2. Brewer grating position. changed @ fixed laser wavelength, dispersion approx. by a quadratic function

3. Brewer grating pos. changed @ fixed laser wavelength, dispersion approx. by a cubic function

4. Brewer grating pos. changed @ fixed lamp emission wavelength, dispersion approx. by a quadratic function

5. Brewer grating pos. changed @ fixed lamp emission wavelength, dispersion approx. by a cubic function

5 Conclusions

- 1. Using the measured slit functions instead of the [..¹⁴⁹]paramterized ones increases the ozone absorption coefficients and consequently the calculated ozone values by 0.8%.
- 2. The quadratic dispersion relation fit used in the standard Brewer algorithm is not suitable outside the ozone spectral
- range 310 nm 320 nm. The residuals show a systematic pattern, which is particularly important [..¹⁵⁰] in the higher end of the spectral range.
- 3. The comparison of the results of the three experiments shows a maximum difference of 0.3% if the cubic fit is used to approximate the dispersion relation of the Brewer instrument. The respective [..¹⁵¹]error in the ozone absorption coefficient [..¹⁵²]obtained using direct measurement in ozone mode and [..¹⁵³]operative discharge lamp method is only of 0.1%[..¹⁵⁴]. This confirms the standard procedure used for the RBCC-E calibrations.

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^{4. [..&}lt;sup>155</sup>]

¹⁴⁹ removed: parametrized

¹⁵⁰removed: at the upper

¹⁵¹removed: difference between

¹⁵²removed: that is obtained from the direct measurements of the tunable laser in the

¹⁵³removed: from the

¹⁵⁴removed: , if both use the parametrized or measured slit

¹⁵⁵removed: This work validates the current wavelength calibration method of the Brewer network and shows that in general there is no need for the characterization of the network Brewer instruments with a tuneable source. However, the limitations of the quadratic dispersion approximation used over extended spectral range of Brewers MK-III and MK-IV are evident. Thus, we suggest to update the operating Brewer software for this model instruments with the recent version, which already includes the cubic dispersion.

$[..^{156}] [..^{157}] [..^{158}] [..^{159}] [..^{160}] [..^{161}] [..^{162}] [..^{163}] [..^{164}] [..^{165}] [..^{166}] [..^{167}] [..^{168}] [..^{169}] [..^{170}] [..^{171}] [..^{172}] [..^{173}] [..^{173}] [..^{171}] [..^{171}] [..^{171}] [..^{172}] [..^{171}] [..^{171}] [..^{172}] [..^{171}$

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- $^{160}\mathrm{removed}:lamp_quad^4$
- 161 removed: $lamp_cubic^{5}$
- ¹⁶²removed: SGW
- ¹⁶³removed: 0.3409 ¹⁶⁴removed: 0.3442
- ¹⁶⁵removed: 0.342
- ¹⁶⁶removed: 0.3446
- ¹⁶⁷removed: 0.3412
- ¹⁶⁸removed: ratio
- ¹⁶⁹removed: 1

 $^{^{156}}$ removed: Ozone absorption coefficient in atm cm^-1 calculated using four absorption cross sections

 $^{^{157}}$ removed: brw_scan^1

¹⁵⁸removed: *opo_quad*²

 $^{^{159}}$ removed: opo_cubic^3

¹⁷⁰removed: 0.9881

¹⁷¹removed: 1.0033

¹⁷²removed: 1.0108

¹⁷³removed: 1.001

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- 10

Wavelength calibration of Brewer spectrophotometer using a tunable pulsed laser and implications to the Brewer ozone retrieval

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We thank the referee for their detailed comments on our manuscript. In the attached document we address their questions and suggestions.

Comments by Reviewer #1

J. Gröbner (Referee) julian.groebner@pmodwrc.ch The paper describes a very detailed and unique characterisation experiment

- 5 of a Brewer spectrophotometer to determine its spectral characteristics (wavelength scale and spectral resolution) which is necessary to calculate the ozone absorption coefficient required for the total column ozone determination from the solar irradiance mea- surements. The experiment was performed using a tunable laser source to compare and validate the standard procedure used by the Brewer community. The results show that the two procedures provide consistent results to within 0.1% which is very satisfying and confirms that the current standard procedure is valid. Apart from minor grammatical errors the
- 10 manuscript is well written and certainly inter- esting to the scientific community. I have a few comments which I would the authors to answer, pending those I support the publication of the manuscript.

page 6, line 10: It is a cubic polynomial fit, not cubic spline.

corrected

- page 7, lines 13-17: It is true that the hg test of the Brewer is repeated when the discrepancy between the actual and determined position is larger than 1.5 steps. How- ever the hg routine sets the position of the micrometer according to the calculation, and repeating the hg routine serves mainly for confirmation. Therefore the hg routine is accurate to +-0.5 steps, since this is the resolution of the system, not +-1.5 steps as written in the manuscript. This considerably improves the estimated wavelength uncertainty. Yes this reduces considerably the error, 1.5 steps are an estimation of the maximum error in the EUBREWNET processing, in this network a mercury test is considered failed when is repeated, the ozone observations are discarded between the last test and the failed one, how ever most of the test gives a correction of less than step.

In section 3.1, I would suggest to add some information on the wavelength uncertainty of the tunable laser setup, which will affect the Brewer wavelength dispersion. I expect in fact the Brewer wavelength dispersion to have less uncertainties when using spectral discharge lamps with published emission line wavelengths ($\approx 1pm$), than the wavelength obtained by the tunable laser system ($\tilde{1}0$ pm).

The text in Section 3.1 was expanded to explain the wavelength calibrations of the instruments used to monitor the laser wavelengths. As to the uncertainty of the Brewer calibration using emission lines, one has to consider also the effect of varying ambient conditions (pressure, temperature, humidity, CO2 content) on the refractive index of air, which is also in the range of several picometers (e.g. ambient temperature change from 20 C to 25 C has an effect of 2 pm, atmospheric pressure change from 101kPa to 98 kPa would cause a 3pm change). So that the uncertainties in both cases should be indeed quite comparable.

- In section 3.1.2 the authors compare the ozone absorption coefficient calculated with the parametrized and the actual slit functions and show that the difference is of the order of 0.9% (Table 2). The parametrized slits however are trapezoidal, with a plateau at 0.87 (13% from the top). However as shown in Figure 6, this is not representing the true slits, and therefore the parametrization might be closer to reality when using the full triangle as parametrization. This might show that the method using the parametrization with a full triangle will have less differences to the tunable laser results using the actual measured slit functions. (I have made some tests and the full triangle parametrization resolves about half of the 0.9% discrepancy). I would suggest that the authors add a third column in table 2 showing this information.

10

Thanks for the suggestion, a new column were added and the calculation repeated, we found the discrepancy is more near to 0.85% +/- 0.1 for the high resolution cross sections rather than 0.9% and the use of the triangular parametrization in fact reduce the difference to the half 0.45%. The results slightly differ from cross section to cross section with the exception of t Bass&Paur probably due his low spectral resolution (Table 1).

2

Table 1. Ozone absorption coefficient in atm cm^{-1} calculated using four absorption cross sections. And the percentage difference to the measured to the paramerizated with a trapezoid and triangular

Trapezoid	Triangular	Measured	% Trapezoid	% Triangular
0.3381	0.3395	0.3406	-0.73	-0.33
0.3331	0.3344	0.3359	-0.85	-0.44
0.3483	0.3498	0.3514	-0.86	-0.45
0.3393	0.3407	0.3422	-0.84	-0.43

- In the conclusion, page 13, last sentence, I do not understand the statement saying that both methods agree to 0.1% if the parametrized or measured slits are used. To my understanding, the standard method using a scanning grating is not able to use the measured slits, since the method relies on interpolating the slit functions to the ozone position, which therefore requires a parametrized slit.

- In my opinion the abstract should also mention the positive result that the tunable laser and the scanning grating method give the same ozone absorption coefficients (to within 0.3% or so)?

The comment were added

1 Minor comments

5 :

- The different wavelength scales (nanometer, angstroem) used in the manuscript and the figures is confusing, and I would recommend to use a single one (nanometer)?

Following your suggestion, we have modified the Figures and, now, its scale is in nanometers. In the text we have tried to express all the results in nanometers. However, some results were expressed as Angstroem or picomentros, because it is a very small value. (Also, the other referee has indicated that some results may be better expressed in picometers) -page 4, line 8 : i would explicitly state that the method is an ozone calibration (not to be confused with a radiometric irradiance calibration for example).

revised.

page 5, point 3: The FWHM also depends on wavelength, which therefore requires some sort of parametrization of the slit function when using the standard scanning grating method.

revised.

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- Figure 2: The units on the left axis seem too small (maximum of 7 counts/second)?
```

5

The exponential were missing, now is corrected.

```
- page 6, line 3: I would remove the value in parenthesis (0.0080 nm), or replace the picometer values.
```

The value has been removed in the text.

-Table 3, I did not find the acronym for SGW. Could it be added in the caption of the Table, for clarity?

10 The SGW makes reference to the article Weber et al. (2016). In this paper, the authors studied the ozone absorption cross section and its uncertainty.

2 initial review

Page 1, Background: Some references on the brewer spectrophotometer should be included.

References to the brewer instrumet are added.

I have never seen such an equation to determine the ozone absorption coefficient within the beer-Lambert law. I even think that it is a circular argument, because the solar spectrum would be included twice, one time directly in the Beer-Lambert Law, the second time for the convolution. Therefore I think this equation is in fact wrong, and the correct one is the one mentioned later (e.g. equation 8. I would suggest that either the authors can provide a reference for this usage of Equation 7, or delete this part of the discussion, which is not used anyway.

Simmilar equation were used in a tutorial by Davd Wardle during the Brewer workshop,

Wardle, D.: Physical Principles II: Optical Characteristics of the Brewer, in The Tenth Biennial WMO Consultation on Brewer Ozone and UV Spectrophotometer Operation, Calibration and Data Reporting, World Meteorological Organization. [online]

5 Available from: "ftp://ftp.wmo.int/Documents/PublicWeb/arep/gaw/gaw176_10thbrewer.pdf" 2008

and on the Thesis Savastiouk (2005) page 94, eq 4.12

Is a simplified reformulation of the more complex equation of Vernier-Wardle (Vanier and Wardle, 1969) used on the Dobson by Bernhard et al. (2005) (Eq 7)

$$\alpha(\lambda) = \frac{\log\left(\frac{\int E_o(\lambda)S(\lambda,\lambda_i)10^{-\alpha(\lambda)X\mu-\beta(\lambda)}\frac{P}{P_o\nu}d\lambda}{\int E_o(\lambda)S(\lambda,\lambda_i)10^{-\beta(\lambda)}\frac{P}{P_o\nu}d\lambda}\right)}{X\mu}$$
(1)

10 This references were added to the article:

Page 6 line 10, Figure number is missing

corrected

Page 7, lines 1-8: I think there is a mistake in the discussion in the precision of the hg test: The HG test allows the wavelength setting to be set to within 0.5 micrometer steps, not 1.5. Then, since one step equals (approximately), 0.0075 nm, 0.5 steps would correspond to 0.00375 nm, or 0.0375A.

Yes there was a mistake, it should be 0.866 steps not Å. The Hg test allows indeed the wavelength setting to be set within 0.5
micrometer step. However, the correction is only applied if the difference is greater than 2 steps. According to the Brewer SOP :
"Corrections to the micrometer position are made, and if the adjustment required is greater than 2 steps (.012nm) then the scan is repeated. "So that we assume that the maximum error is +-1.5 steps with a rectangular probability distribution function. Hence, the standard uncertainty in such a case is 1.5/sqrt(3) = 0.866 steps, which is approx 0.065Å (0.0075 nm per step)

Figure 5. Why use of log scale? In this manuscript, the interest lies in the slit function shape, and therefore in the linear scale, not in the wings.

The reason to use log scale is to visualize the dark count issue, in linear scale is difficult to see.

page 10, last line. I would add "as already noted in Gröbner et al., 1998", at the end of the sentence finishing with (310-320 nm).

5 added.

I am confused by the conclusion, and would suggest that the authors rephrase it be less ambiguous: Point 1 of the conclusion states that the tunable laser results give an ozone absorption coefficient 0.8% higher than the ones obtained with the standard approach using the parametrized slits. The last point states however that the use of the cubic spline agrees with the tunable laser results to within 0.1% and therefore confirms that the use of the tunable laser (fixed grating position), with the standard procedure (rotating grating) is equivalent. While I understand that cubic and quadratic dispersion fittings might give rise to differences, I do not understand how this affects the use of parametrized slits and actually measured ones using a tunable laser.

We rephrase the conclusions, to reflect from one side the use of parametrized slit function gives a significant difference of 0.8% if we use the measured slit, but the use of the laser confirm the operational rotating grating method (using the parametrized slits with the laser)

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Vanier, J. and Wardle, D.: The effects of spectral resolution on total ozone measurements, Quarterly Journal of the Royal Meteorological Society, 95, 399, 395, http://adsabs.harvard.edu/abs/1969QJRMS..95..395V, 1969.

Weber, M., Gorshelev, V., and Serdyuchenko, A.: Uncertainty budgets of major ozone absorption cross sections used in UV remote sensing ap-

10 plications, Atmospheric Measurement Techniques, 9, 4459–4470, https://doi.org/10.5194/amt-9-4459-2016, https://www.atmos-meas-tech. net/9/4459/2016/, 2016.

Wavelength calibration of Brewer spectrophotometer using a tunable pulsed laser and implications to the Brewer ozone retrieval

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We thank the referee for their detailed comments on our manuscript. In the attached document we address their questions and suggestions.

Comments by Reviewer #2

1 General comment

- 5 The paper describes an experiment of a spectral characterization of a meteorological instrument: The Brewer (principally used for UV radiation and total ozone column monitoring). This experimentation has been done in a laboratory of a metrological institution (the German PTB in Braunschweig) with the help of one of the most modern and precise material: A tunable laser. The originality of this experiment (part of the EMRP/ATMOZ project), is that scientific teams of the meteorological community worked together with metrology specialists, what is a unique gain for the quality management of the meteorological
- 10 measurements. The spectral characterization that has been done is necessary for the determination of the total ozone column from the UV irradiance measured with the instrument Brewer. The results of the experiment allow a high quality validation of the standard procedures of spectral characterization used in the Brewer community, and allow defining quantitatively the limits of these standard procedures and of their assumptions. The results are well presented in this article, and the discussion is of high scientific quality and will be useful for the Brewer community.
- Apart from minor (but many) typology and grammatical errors, that should be corrected (I have listed a part of them at the end of this report), the manuscript is well written pleasant to read and well understandable. I have a few (5) comments/questions which I would the authors to answer, pending those I support the publication of the manuscript. Technical comments ("x-y = page x, Line y):

5 Comments/questions about the content:

1) In the beginning of the background paragraph, when you mention the slits of the "slit mask" (2-1), it would be welcome to have a brief description of the brewer's slits, and to what they are useful. Maybe you should introduce a table of them and refer to the table in the text.

A table with the slits (Number and its wavelength) and a description has been added in the text.

2) (in 1. Background), lines 2-10 to 2-15, you mention some assumptions about the slit functions (2-14). It would be nice to explain before or in a table, what are the most important assumptions that are done.

The assumptions are described later, we add a reference to the section on the text.

3) In the description of the experiment (Background from 2-19 to 2-28): 3a) I guess the use of the tunable laser is useful for point 2, not point 3. So the mention "using the tunable laser" (2-24) should go in the title of point 2 (2-20). yes 3b) In point 3, you mention that the Brewer scans +-2nm around a fixed wavelength of the laser. Further in Point 3, you mention that the Brewer can scan with 5nm increment between 290 nm and 365 nm. You need to precise how you can scan with a lower step than the increment.

5

Yes is an error, the experiment were performed with the laser emitting from 290 to 355 every 5 nm whereas the brewer is scanning every 5 steps.

4) In the alpha-formula (4-24) you write the ozone cross-section with alpha(lambda) and then in the text, you mention sigma for it.

Corrected

5) In the discussion, it would be welcome to explain how the different Brewer users can use these results to optimize the TOC retrieval with their own Brewer. Can these results be generalized to all Brewers? To all Mk-III Brewers? Or should every Brewer go to a laboratory with a tunable laser to characterize its slits? Do you have assumption concerning the age stability of this slit characterization? Is it expected to change with the time? If yes why?

10

2

The work validate the method currently in use with the brewer network, the limitations of the quadratic dispersion is an issue on MK-III and MK-IV brewer and the new operating software recently introduce the cubic dispersion. We don't think necessary the characterization with the laser of network brewer.

We introduce this on the discussion.

5 Grammar and typos

You use sometimes "tuneable", sometimes "tunable", please choose one terminology, and I guess "tunable" is the correct one. -> So please correct in title, in 1-5 (Abstract), in 2-9, 2-13 and 2-16 (Chapter 1), in 7-22 and 8-1 (Chapter 3) **Done**

Abstract

1-9: "a underestimation" -> an underestimation Done

10 1.Background

1-13: "total column ozone (TOC)"-> total ozone column (TOC) 2-9 "the use of the use of" -> the use of 2-9 "allow us" ->

allows us Done

2-10 "coefcients" -> coefficients **Done**

- 2-14 "the need for the assumptions" -> the need of assumption **Done**
- 15 **2.** Calibration of the Brewer sprectrophotometer 3-7 "weighting coefficients w" -> "weighting coefficients wi" Done
 - 5-11 "referered" -> referred **Done**
 - 5-12 "double Brewer," -> double Brewer, (without space before the ",") Done

6-1 "one of the six exit slit" -> one of the six exit slits **Done**

6-7 "0.7 A" -> 0.7 Å (A with "o" on the top for Ångström) **Done**

20 7-1 "The cubic approximation method. . . use knowledge" -> ". . . uses . . ." Done

3. Pulsed laser-based measurements

8-10 and 8-11 "counts/seconds" (3 times)-> counts/second Done

9-7 "parametrised" -> parametrized Done

9-8 "(Orphal et al. (2016)." -> ", Orphal et al. (2016)." or (Orphal et al. (2016)) Done