TOC intercomparison of Brewer, Dobson and BTS Solar at Hohenpeißenberg and Davos 2019/2020

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Abstract. In the 2019/2020 measurement campaign at Hohenpeißenberg (Germany) and Davos (Switzerland) we compared the well-established Dobson and Brewer spectrometers (single and double monochromator Brewer) with newer BTS array spectroradiometer based systems in terms of total ozone column (TOC) determination. The aim of this study is to validate the BTS performance in a longer-term TOC analysis over more than one year with seasonal and weather influences. Two different BTS setups have been used. A fibre-coupled entrance optics version by PMOD/WRC called Koherent and a diffusor optics which proved to be simpler in terms of calibration from Gigahertz-Optik GmbH called BTS Solar. The array-spectrometer based BTS systems have been traceable calibrated to National Metrology Institutes (NMI) and both versions of TOC retrieval algorithms are based on spectral measurements in the range of 305 nm and 350 nm instead of single wavelength or wavelengths pair measurements as for Brewer or Dobson. The two BTS based systems, however, used fundamentally different retrieval algorithms for the TOC assessment, whereby the retrieval of the BTS solar turned out to achieve significantly smaller seasonal drifts. The intercomparison showed a difference of the BTS Solar to Brewers of <0.1% with an expanded standard deviation (*k=2*) of <1.5% within the whole measurement campaign. Koherent showed a difference of 1.7% with an expanded standard deviation (*k=2*) of 2.7% mostly caused by a significant seasonal variation. To summarize, the BTS Solar performed at a level of Brewers at the comparison in Hohenpeißenberg. The slant path slope is in-between double monochromator and single monochromator Brewer. Koherent shows a strong seasonal variation in Davos due to the sensitivity of its ozone retrieval algorithm to stratospheric temperature.

1 Introduction

Atmospheric ozone is recognized an essential climate variable in the global climate observing system (GCOS) of the WMO. Careful long-term monitoring of the global ozone layer is still crucial in verifying the successful implementation of the Montreal Protocol and its amendments (MPA) on the protection of the ozone layer, with the eventual recovery of the ozone layer to pre-1970's levels.

McKenzie et al. (2019) showed for instance that no global, statistically significant recovery of the ozone layer took place yet, but no further decline was either observed. This alone suggests that longer periods of observation are necessary. In addition Montzka et al. (2018) stated that despite the ban, CFC-11 has been released in measurable quantities in China. This means that not only the control of the expected recovery of the ozone layer, but also the monitoring protocol for the CFC ban are important tasks (Bais et al., 2019).

On the other hand, the effects of the global climate change to the ozone layer are yet not completely understood (Bais et al., 2019;Bais et al., 2018;Bais et al., 2015;Seckmeyer et al., 2018) which is an requirement for continuing observations.

The monitoring of the ozone layer started already in 1926, with the development of the Dobson Spectrophotometer designed and built by Prof. G.M.B. Dobson at the Oxford University (Dobson, 1931;Dobson, 1968). The first small network with six stations (Abisko, Arosa, Lerwick, Lindenberg, Oxford, Valentia) was established in 1926 (Köhler and Claude, 2006), to measure the total ozone column (TOC). A remarkable extension of the global network took place since 1957/1958

 (International Geophysical Year). In the following decades, up to 130 instruments were produced and employed. The Meteorological Observatory Hohenpeißenberg (MOHp) of the German National Weather Service (DWD) started its observations of the ozone layer with ozone sonde (vertical profile of the ozone up 35 km) and with Dobson No. 104 (TOC) in 1967/68. Approximately 50 stations with Dobson instruments are still operational today and submitting data to the World Ozone and UV Data Centre (WOUDC) in Toronto (Canada). Thus, this type of spectrophotometer is still a backbone in the

45 global ozone monitoring network, although new, modern instruments like the Brewer Spectrophotometer have been established in this network since the 1980s.

State-of-the-art array spectrophotometers measuring spectral UV radiation are potential candidates for such new instruments (Egli et al., 2016). These have e.g. the advantage that no wavelength scanning (moving parts) is needed (mechanically less complex systems). Due to strong ozone absorption between 300 nm and 350 nm, TOC can be retrieved from spectral direct

50 sun measurements in the UV band (Huber et al. 1995). The same spectral range is used for TOC retrieval from Brewers and Dobsons (Kerr et al., 1988). However, the advantage of array spectroradiometers is that a spectrum is measured at wavelength step increments <0.2 nm instead of only a few wavelength values as for the Brewer (5 wavelengths) or the Dobsons (single and double pair observations). The larger number of wavelengths is named here as "full spectrum" in order to distinguish from the four or five wavelengths approach of the Dobsons or Brewers. One may assume that this additional spectral information

55 from the full spectrum increases the reliability of TOC retrieval. The general specifications for spectral global solar UV measurements in the TOC relevant band are stated in several guideline-publications (e.g.: (Seckmeyer *et al.*, 2010; Seckmeyer *et al.*, 2001). The performance of global UV measurements of different commercially available array-spectroradiometer have been tested at the intercomparison presented in Egli *et al.*, 2016. The results of the global UV intercomparison in Egli et al. 2016 showed that the tested instruments suffered significantly from straylight. In particular at high solar zenith angles, the UV

60 radiation at short wavelengths between 300 nm and around 315 nm are significantly biased, while the spectral measurements at longer wavelength are well reproduced. Just at low SZA some well characterized and calibrated devices provided acceptable deviation in solar irradiance in the range of 5% to the reference.

First instruments using the full spectrum of array spectroradiometer for TOC retrieval are Pandora (Herman et al., 2015) and Phaeton (Gkertsi et al., 2018). The Pandora system retrieves TOC by spectral fitting of the attenuated spectrum in the

65 wavelength band between 305 nm and 330 nm. Herman et al. (2017) reported an averaged difference of 2.1 ± 3.2% for TOC with a relative drift of 0.2% per year of the Pandora system No. 34 compared to the NOAA Dobson No. 061 in Boulder Colorado based on a three years intercomparison. For the comparison, both instruments have been corrected for the impact of stratospheric temperature using their standard ozone absorption cross section. The Dobson was not corrected for straylight. However, AD-pair direct sun Dobson observations were used in comparisons with Pandora within the acceptable range of air

70 masses that would minimize the impact of straylight. Additionally, a correction has been implemented for the Pandora system addressing the effect of straylight.

The Phaeton instrument is a DOAS/MAXDOAS system using the wavelength band between 315 nm and 337 nm. The instrument was characterized for straylight with a tuneable laser and a straylight correction was applied. The two-years comparison with a single monochromator Brewer spectrophotometer revealed an average bias of 1.85 ± 1.86% for TOC using

75 the Paur and Bass (1985) cross-section at constant temperature of 228K. When including the daily variability of the stratospheric temperature the differences are reduced to 0.94 ± 1.26%.

The BTS array spectroradiometer has been released after the assessment of quality of aforementioned global UV array spectroradiometers (Egli et al., 2016). The accuracy and stability of BTS solar global spectral irradiance measurements were compared against well-established double monochromator-based systems (Zuber et al., 2018a). The BTS reduces straylight by

80 filtering optical radiation from longer wavelength with different bandpass filters inside the array-spectroradiometer. Due to this straylight suppression, the UV index measurents derived from the BTS spectra were within ±1% for solar zenith angles (SZA) smaller than 70° and ±3% for SZA between 70° and 85° in reference to an NDACC device (Zuber et al., 2018a). The

comparison of the BTS solar UV spectra with the reference showed some biases of up to 5% for wavelengths shorter than 310 nm for SZA larger than 70° (Zuber et al., 2018a). These biases may affect the TOC retrieval using the full spectrum, including

85   short wavelengths. Addressing this question, Zuber *et al.*, 2018b noted a deviation of TOC determination of less than 1.5% (for SZA smaller than 65°) to other instruments in most situations and not exceeding 3% from established TOC measurement systems such as Dobson or Brewer at the Izaña Atmospheric Observatory

in Tenerife. At higher SZA the deviation slightly increased for the TOC determination. This may be an effect of the remaining straylight in the wavelength band between 305 nm and 310 nm, which was used during this intercomparison. Potential effects

90   on TOC caused by the remaining straylight will be further discussed based on the results of his study.

In this publication we show the longer-term (more than one year) performance of BTS based systems concerning TOC determination in terms of different solar zenith angles, different TOC level, seasonal and weather influences in harsh environment at Hohenpeißenberg (Germany) and Davos (Switzerland) for a period of more than one year. Furthermore, two distinct different retrieval algorithms from the BTS Solar and the Koherent system are compared and discussed based on the

95   results of this long term intercomparison.

2 Measurement campaign, instruments and retrieval

The intercomparison took place in the 2019/2020 measurement campaign at the Deutscher Wetterdienst DWD Hohenpeißenberg, Germany (altitude: 953 m; coordinates: 47.80 N, 11.01 E) and Physikalisch-Meteorologisches Observatorium Davos (PMOD/WRC), Switzerland (altitude: 1580 m; coordinates: 46.81 N, 9.83 E).

100   Both stations are part of the Global Ozone Monitoring Network of WMO/GAW (World Meteorological Organisation / Global Atmosphere Watch) and the European Brewer Network (http://www.eubrewnet.org/). The DWD Hohenpeißenberg has been the WMO Regional Dobson Calibration Centre for Europe (RDCC-E) since 1999 and is part of the GAW-network since 1995 (Köhler, 2002). The PMOD/WRC is the world calibration centre for meteorological radiation measuring instruments. At Hohenpeißenberg the BTS Solar is compared to the instruments Dobson No. 104, Brewer No. 010 (single monochromator)

105   and Brewer No. 226 (double monochromator). At Davos the BTS based Koherent is compared to the Brewer No.163 (double monochromator).

2.1 Dobson/Brewers

Today Dobson and Brewer spectrophotometers are still the main instruments in the global network used to monitor the ozone

110   layer, and have been in use since the 1920's and 1980s respectively.

The operational instruments, used at the Hohenpeißenberg Observatory are one field and one reference Dobson spectrophotometer and two field Brewer spectrophotometers, both types of spectrophotometer use a similar physical principle. They measure the incoming solar radiation in various spectral lines in the short UV-B (280 – 315) nm and the longer UV-A range (315 – 400) nm. Whereas the short wavelengths are affected by ozone absorption, the longer wavelengths longer than

115   340 nm are unaffected by ozone absorption.

The Dobson spectrophotometer measures the relative intensity of the solar radiation at three wavelengths pairs, using a photomultiplier tube (PMT) and a so-called optical wedge. Detailed descriptions of the Dobson design and function, the data processing algorithms and the treatment and minimizing of special instrumental and atmospheric effects (internal and external straylight like mie scattering, dependence on the effective temperature of the ozone layer etc.) can be found in a large plurality

120   of old, historical and recent literature. Dobson relevant papers are: Dobson (1931), Dobson (1957a), Dobson (1957b), Dobson (1968), Komhyr (1980), Evans (2008), Staehlin et al. 2003 and Vogler et al. 2007. .

The measurement principle of the Brewer spectrophotometer is similar, as it uses the spectral lines of the solar radiation in the same UV-range as the Dobson. As the Dobsons, the TOC retrieval with Brewer is obtained with the double ratio technique of the wavelength pairs using a slightly different weighting for the individual channels. Publications describing the Brewer Spectrometer are(Kerr et al., 1985;Kerr et al., 1981). The essential difference is that this spectrophotometer measures the photon counts directly with a photomultiplier in each of the used five wavelengths for ozone and additionally SO_2. The same procedure of the double ratio technique as with the Dobson is then applied to derive the TOC. One advantage of the Brewer type is its automatic operation using a PC with a special control and TOC-calculation software, which is in contrast to normal Dobsons, that are manually operated (except the Swiss automatic Dobsons, Stübi et al. (2017)). However, both methods of measurements, manual or automatic, need several minutes (Brewer e.g. allow a single measurement within 30 seconds, however typical 3 minutes are needed in order to detect cloud influences) to obtain a reliable value of the TOC.. At this point, it should be noted for comparison that the fully automated BTS based devices allow reliable measurements within 8 to 45 seconds (depending on SZA and BTS setup), however usually an averaging of 1 min to 5 min is applied in order to receive a practical the amount of data and optimize the signal to noise ratio.

Many publications about medium and long term intercomparisons between both spectrophotometer types have been published in the past three decades: Kerr et al. (1988), Köhler and Attmannspacher (1986), Köhler et al. (1989), Staehelin et al. (2003) with a detailed description of function and ozone retrieval algorithms of Dobson and Brewer, Scarnato et al. (2010). An important publication on the compatibility of Brewer measurements in Arosa and Davos have been published by Stübi et al. (2017). The paper of Redondas et al. (2014) describes the effects of the planned introduction of new ozone absorption cross sections (Gorshelev et al., 2013;Serdyuchenko et al., 2013) on the Brewer and Dobson ozone retrievals. The comparison of Dobsons and Brewers with different cross-sections is recently performed at PMOD/WRC (Gröbner J., 2021). In addition to the introduction of new ozone absorption cross sections a further potential improvement of Dobson data is the use of effective absorption coefficients for each Dobson. The determination and application of these individual absorption coefficients, taking into account the effective temperature of the ozone layer and individual effective slit functions for each Dobson, has recently been investigated and discussed (Köhler et al., 2018;Redondas et al., 2014).

The last official calibration services of the Dobson No. 104 with the Regional Standard Dobson No. 064 of the Hohenpeißenberg RDCC-E were performed in 2018 and 2019 and confirmed its stable calibration level, as the agreement with the standard was better than 1%. The Hohenpeißenberg Brewers No. 010 (running since 1983) and 226 (running since 2014) have undergone a calibration service in July 2020, done by IOS using the traveling standard Brewer No. 017 (see Figure 2). These kind of calibration services have been performed every year in the past 20 years. The stability of the calibration levels of both Brewers (differences to the reference <1%) have been confirmed almost every year, so in 2020 too.

Both systems are still in regular use, even though Dobson spectrophotometers are no longer being manufactured. In order to further maintain a dense network of TOC observations worldwide, new automatic, reliable, simple and cost-effective systems have been developed.

2.2 BTS Solar

BTS Solar is based on a BTS unit with directly mounted diffusor entrance optic and a tube for field of view FOV limitation for irradiance measurements (no optical fibre). The device is described and characterised (e.g. FOV = ±1.4°) in detail in Zuber et al. (2018b). For the TOC determination a least-squares minimization algorithm in the wavelength range from 305 nm to 350 nm according to Huber et al. (1995) could be used to take advantage of the entire full spectrum, as is used by the Koherent measurement system discussed in section 2.3. However, for the BTS Solar, and in contrast to Koherent, we developed a different TOC retrieval algorithm, which long-term/seasonal performance is validated in this study. . The algorithm is also described in more detail in the Zuber et al. (2018b). In summary, the technique is based on the comparison from two selected

wavelength bands (instead of the entire spectrum as for Koherent) of the measured data with a lookup table (LUT) pre-calculated by the libRadtran software package for radiative transfer calculations (Emde et al., 2016).

For the intercomparison within this paper two adaptions to previous campaign retrieval algorithm (Zuber et al., 2018b) have been applied.

170 First, the two wavelength-bands which are considered for the TOC retrieval are adapted to 307 nm to 311 nm (high ozone absorption) and 319 nm to 324 nm (low ozone absorption, see Figure 1), which showed to be the most robust configuration for the BTS device. Reason for this choice is that both wavelength bands are within one optical filter measurement of the BTS and the measurement time could be further decreased in principle to this single filter measurement. Second, an air pressure correction for this retrieval algorithm has been introduced in order to consider the larger differences that can be expected within

175 a period of the measurement campaign. A correction model has been used since the calculation of a different LUT for every air pressure would be possible but it is too calculation-time extensive. The standard deviation ($k=1$) of the correction within the whole expected air pressure variability at Hohenpeißenberg is <0.8 DU, which has been derived by comparing the model with calculated LUTs for different air pressure (for an air pressure range of 870 hPa to 930 hPa, as observed at Hohenpeißenberg, a correction of maximum 2.5 DU results). For this purpose, five different LUTs with different pressures

180 where calculated and a best fit model for the pressure correction was determined. This correction has been applied after the TOC retrieval based on a standard pressure with the actual present air pressure during the measurement campaign. The air pressure data has been supplied by the DWD.
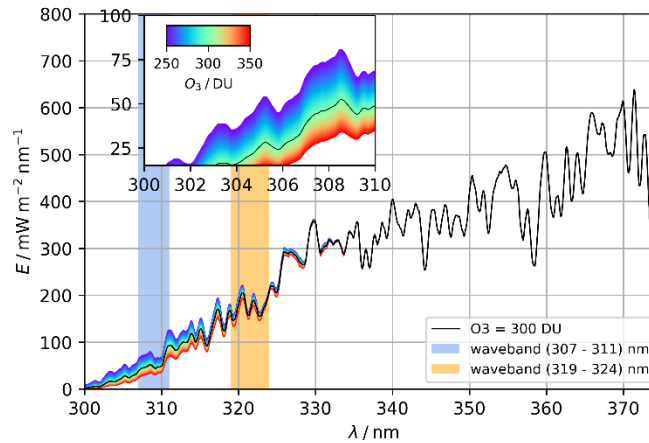
Figure 1: Direct irradiance spectra of the lookup table modelled with libRadtran. The two wavebands for the TOC retrieval are
185 **marked with blue (high O3 absorption) and orange (low O3 absorption)**

In general, for the calculation of the LUT, the following values for the input parameters have been chosen for Hohenpeißenberg: air pressure as stated above, an altitude of 953 m, an atmospheric profile typical for subarctic (due to the mountain region) summer and winter (transition in April and October) given by Anderson et al. (1986), the ozone cross section of Paur and Bass (1985) (the IUP cross-sections of Serdyuchenko et al. (2013) are in the meantime also available for libRadtran.

190 A sample check of one day (24.02.2020) results in a difference of 0.4 DU when using Bass and Paur or IUP. Finally, the high resolution QASUMEFTS extra-terrestrial spectrum of Gröbner et al. (2017) is used for the libradtran modelling. For aerosol, albedo (which has no significant influence to direct solar irradiance) and SO_2 standard values of libRadtran have been used. This methodology was applied with excellent results to measurements at Izaña Atmospheric Observatory in Tenerife (Zuber et al., 2018b) using slightly different wavelength for the ozone retrievals. As well at the intercomparison in Huelva with

195 comparable results which have not been scientifically published yet. The results of this study with the adopted wavelength show that a retrieval based on a comparison of the LUT to the measurement provided accurate results for many cases. At this point we want to emphasize that this retrieval method based on LUT does not require any further time dependent input parameters apart from the current air pressure. However, a more precise and extensive modelling also for each measurement individually, would be possible and maybe also needed for some measurement sites with different and fast changing

atmosphere. However, in the libradtran software package further atmospheric profiles are available and can be applied for other sites (midlatitude summer and winter, subarctic summer and winter, tropical and US standard atmosphere) (Emde et al., 2016). During this campaign, the mentioned pre-defined standard parameters were used for the LUT and were not changed within the whole intercomparison campaign except for the seasonal atmosphere model (summer/winter), which was changed in April and October. At the date of the season change no significant change in TOC retrieval was recognizable at the evaluated air mass factors up to 3.5. No additional correction of stratospheric ozone temperature has been applied so far.

The BTS Solar has been calibrated for this campaign with a well-known spectral irradiance calibration setup in 70 cm distance at the Gigahertz-Optik ISO 17025 certified calibration laboratory (D-K-15047-01-00, D-PL-15047-17025-2020). This calibration showed to be robust and small measurement uncertainties of 2.5% (expanded measurement uncertainty k=2) in spectral irradiance can be achieved (Zuber et al., 2018b;Vaskuri et al., 2018). After 6 month a check of the calibration has been performed, which showed no significant change (differences below the calibration uncertainty of the standard lamp). Resulting the calibration has not been changed within the whole measurement campaign.

A measurement interval of 2 minutes has been chosen. During the whole measurement campaign at Hohenpeißenberg, only 16 days were affected from power breakdowns due to construction work at the site and an IP server issue/software conflict in the beginning of the campaign. These are about 3% downtime during the whole campaign. For the performance evaluation all measurement data of the whole intercomparison campaign were considered. Only measurements with a standard deviation error of 10 DU within five consecutive measurements have been removed since such a large change in TOC within such a short time interval can only be expected due to instrument malfunction, or cloud movement or very high SZA.

2.3 Koherent

Koherent is based on a fibre coupled BTS-2048-UV-S-F array spectroradiometer. Contrary to the instrument at Hohenpeißenberg, the BTS-2048-UV-S-F is connected to a lens-based imaging telescope, with a field of view of about $\pm 0.6°$. The telescope is mounted on a sun tracker on the measurement platform at PMOD/WRC as presented in Egli and Gröbner (2018). The array spectroradiometer is embedded in a temperature stabilized weather-proofed box keeping the instrument at a constant temperature of (22 ± 1) °C through all seasons of the year.

The raw data of the BTS is acquired with a python software-routine on an embedded computer inside the box, providing data with almost no technical failure during the entire campaign. The readings are taken with an interval of 1 min for each measurement. Usually, the device needs around 45 seconds for one measurement. However, at high SZA in the early morning or late evening with low direct sun irradiance intensity, around 2 minutes are needed due to increased integration time. The data was aggregated to a 5 minutes data-point, by averaging the individual data. If the standard deviation of the individual measurements within 5 minutes exceeded 10 DU, the data was considered as invalid. These small-time-scale variations are caused by instable atmosphere such as cirrus clouds or noise from the detector at low sun elevations. The criterion of 10 DU was chosen to remove theses outliers.

The postprocessing was performed off-line with the following steps: a) converting the raw-counts of the readings to irradiance using the laboratory calibration, b) wavelength shift correction using the MatShic software developed at PMOD/WRC (Egli, 2014) and c) retrieving TOC with a least-squares minimization algorithm according to Huber et al. (1995) and Vaskuri et al. (2018).

The aforementioned least-squares algorithm is using a spectral fit in the wavelength range from 305 nm to 350 nm and an atmospheric model based on the Beer-Lambert law:

$$I_\lambda = I_0 \, exp[-\tau_\lambda m] \qquad \text{(Eq.1)}$$

I_λ represents the measured solar irradiance from Koherent at the specific wavelength λ and I_0 is the extraterrestrial spectrum at the top of the atmosphere. For the retrieval, the QASUMEFTS extra-terrestrial spectrum is applied (Gröbner et al. 2017).

The airmass m is important for the attenuation of ozone through the atmosphere, which depends on geographical location and the time of the day and thus on the solar zenith angle during a day and the seasons. . The airmass for the ozone (m_λ^{O3}) , aerosol (m_λ^{AOD}) and Rayleigh (m_λ^R) optical thickness (Eq. 2) is calculated using the different profiles of the standard US atmosphere *afglus* (NOAA, 1976) separately.

$$\tau_\lambda \cdot m_\lambda = \tau_\lambda^{O3} \cdot m_\lambda^{O3} + \tau_\lambda^{AOD} \cdot m_\lambda^{AOD} + \tau_\lambda^R(p) \cdot m_\lambda^R \quad \text{(Eq.2)}$$

. The attenuation is summarized by the term τ_λ, indicating the absorption and scattering by ozone, aerosols and Rayleigh (Eq. 2). The absorption cross section by ozone is provided by University of Bremen, IUP (Serdyuchenko et al., 2013). For the retrieval, the IUP cross-section at the stratospheric temperature of constant -45° is used for the standard US atmosphere. The assumptions of -45°C, the same as used in the standard Brewer procedure.

Finally, the aerosol absorption is parametrized with a spectrally linear model and the Rayleigh scattering by the parametrization of Nicolet (1984) assuming a constant ground air pressure of 820 hPa (for the higher altitude station Davos).

The advantage of the minimal least-square fit approach is that the attenuation by aerosol optical depth is a free fitting parameter for each measured data during the ozone retrieval and is not required as an input parameter.. Note that ozone and aerosol optical depth are weakly correlated, which allows fitting the free variables by the least square fit algorithm. The stratospheric temperature and the height of the stratospheric layer are currently not retrieved by the algorithm as well as SO$_2$ is not included. In summary, the retrieval algorithm used for this comparison, displays a first simple approach to test the performance of the above standard settings of the retrieval method.

The system was calibrated for absolute irradiance at the PMOD/WRC laboratory facilities on 14th and 19th of November 2019 using four 1000W FEL lamps on a linear stage at distances of 2.40 m and 1.30 m. Due to the low light-throughput of the fibre-coupled telescope setup and the sensitivity of the alignment on small angular deviations on the linear stage, the responsivity varied in TOC differences of 2.2% (*k=1*). An averaged responsivity of the system, which preliminary appeared to be the best calibration, was used for the calibration of the raw data and resulting TOC from Koherent presented here.

In analogy to the comparison at Hohenpeißenberg, Koherent is compared with the PMOD/WRC double monochromator Brewer No. 163. The Brewer is annually calibrated for TOC retrieval by the Regional Brewer Calibration Center Europe (RBCC-E) with an uncertainty of around 1% compared to the reference Brewer of the Calibration Center (Redondas et al. 2019), while the stability of the instrument is operationally monitored on a daily schedule. However, an absolute uncertainty of the Brewer No. 163 is not known. Due to this quality and stability assurance of the Brewer No. 163 TOC data, the Brewer No. 163 serves as a reference for the comparison with Koherent in Davos. Brewer TOC readings are taken on an irregular schedule, since the Brewer also measures operationally spectral global UV irradiance.

3 Results of the Intercomparison

3.1 BTS Solar at Hohenpeißenberg

A special day of the intercomparison at Hohenpeißenberg (July 28, 2020) is presented in Figure 2. During this day the RDCC-E reference Dobson No. 064, the Indian Dobson No. 036 and the IOS Brewer No. 017 for service purposes joined the intercomparison. The measurements of seven instruments (BTS Solar, Dobson No. 036, 064 and 104, Brewer No. 010, 017 and 226) showed a good agreement with differences between all instruments mostly less than ±1% (see figure 2) on this special day. Only at low sun elevations, in the morning the BTS solar and the single monochromator Brewer No. 010 show a bias of about 5 DU to the other instruments.
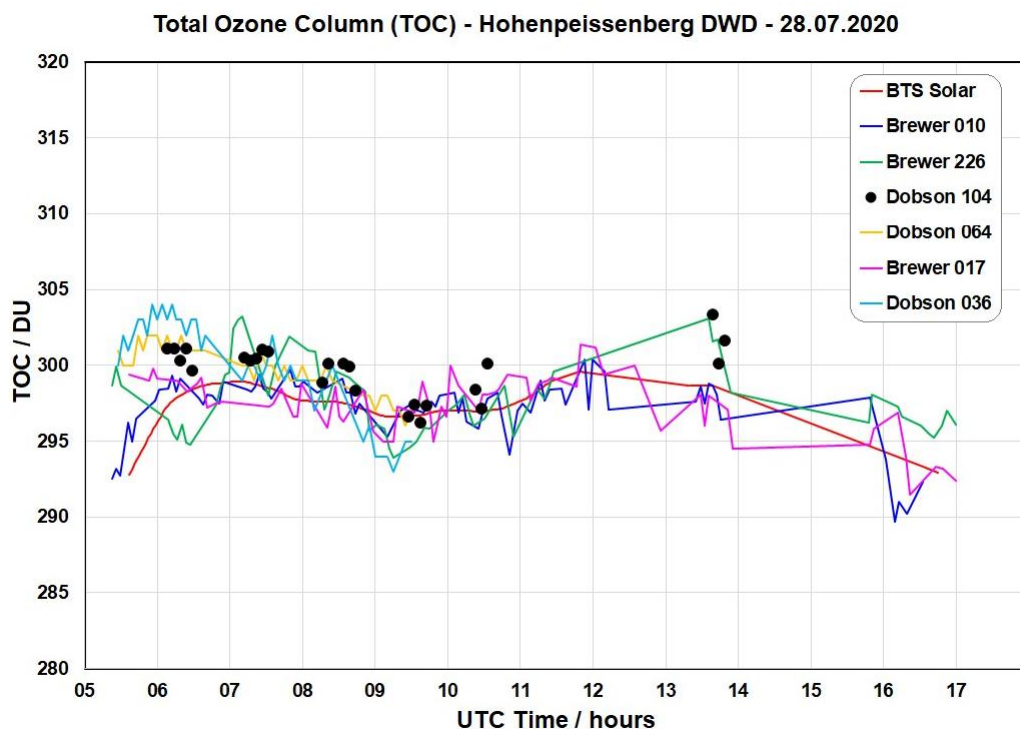
Figure 2 TOC measurements of the IOS Brewer calibration service on the 28.07.2020 at Hohenpeißenberg

In the following, Figure 3 presents two examples of daily courses of TOC. One shows a strong diurnal dynamic and the higher temporal resolution of BTS Solar and Brewers compared to the Dobson. These data show that the BTS Solar and both Brewers capture about the same TOC variability (SZA) The Dobson shows the aforementioned offset in winter season and delivers only a few data points for each day due to its manual operation. However, the Dobson is also able to capture atmospheric changes if a sufficient amount of data is measured (time interval).
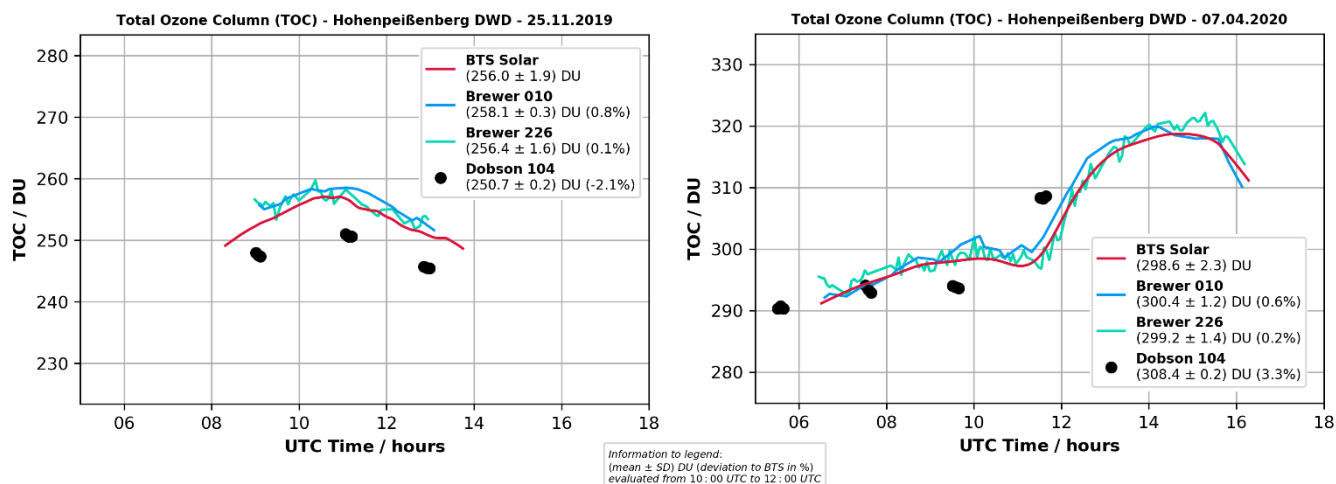
Figure 3 Two exemplarily days of the intercomparison. One shows a strong dynamic in TOC within the day.

Since different devices are compared to the BTS Solar and all of them exhibit their own measurement uncertainty (e.g. 3.7 DU for the BTS, see Vaskuri et al. (2018)) we compared all data in Figure 4 relative to the BTS Solar.

Figure 4 shows that the histogram of all data is comparable between the Brewer Nr. 010 with a mean deviation of 0.04% (percental deviation between the devices (e.g. (Brewer010-BTS Solar)/BTS Solar·100) and an expanded standard deviation ($k=2$) of 1.29% and Brewer No. 226 with a mean deviation of 0.06% and an expanded standard deviation ($k=2$) of 1.47%. The Dobson No. 104 shows a mean deviation of -0.84% and an expanded standard deviation ($k=2$) of 2.22%.
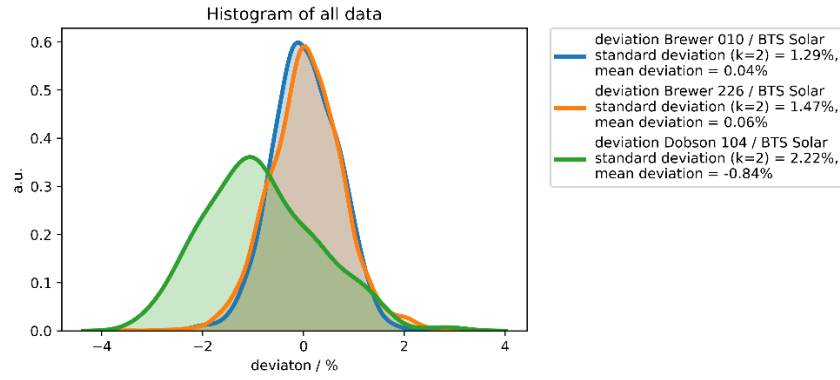
Figure 4 Histogram of all Brewer No. 010, Brewer No. 226 and Dobson No. 104 compared to BTS Solar data from Hohenpeißenberg. The data shows a comparable performance between Brewer Nr. 010 (deviation of 1.29% to BTS Solar), Brewer No. 226 (deviation of 1.47% to BTS Solar) and BTS Solar and a larger mean and standard deviation of Dobson No. 104 (deviation of 2.22% to BTS Solar).

In the following Figure 5 the seasonal course of all TOC data of the complete measurement campaign is illustrated. This explains the larger deviation of the Dobson No. 104 due to the seasonal dependency (Gröbner J., 2021;Kerr et al., 1988;Scarnato et al., 2010;Staehelin et al., 2003;Vanicek, 2006). Figure 5 also shows the comparable results of the Brewers and BTS Solar, since the least-square fit of the full data is within ±1% TOC deviation over the whole measurement campaign.
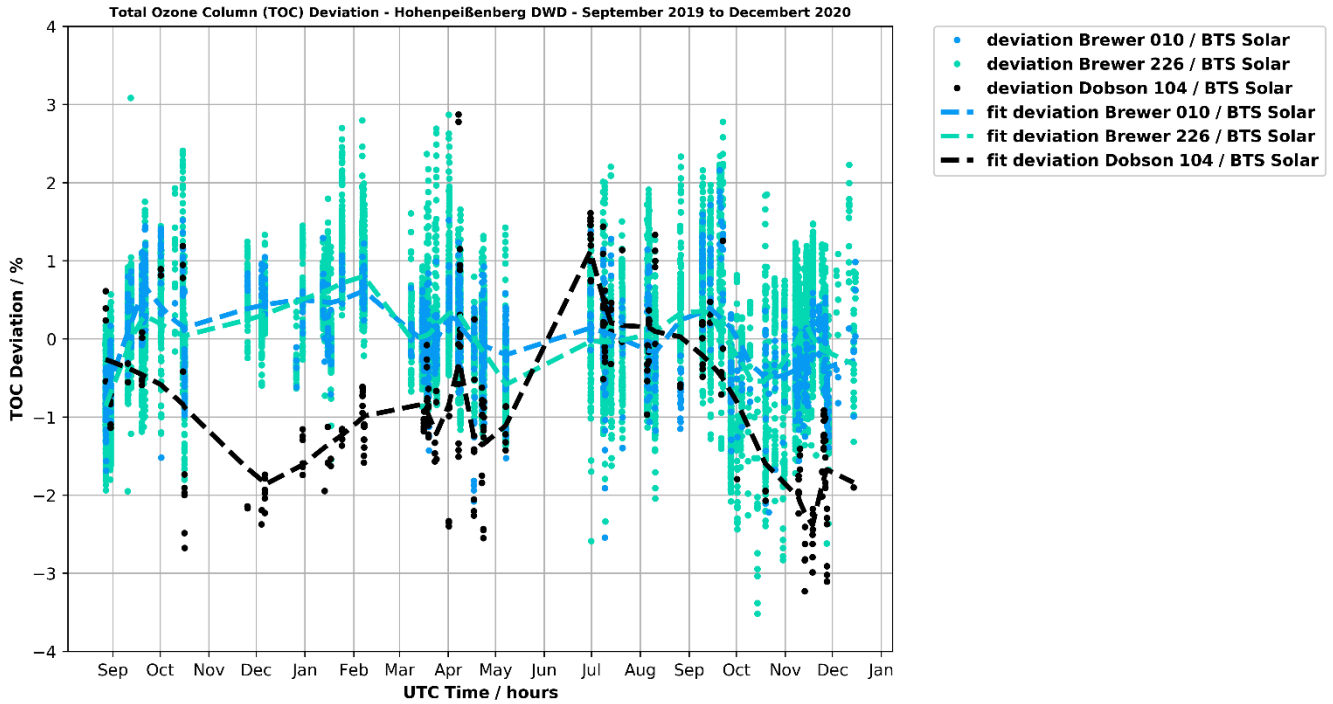
Figure 5 Full measurement campaign TOC plot from Hohenpeißenberg. This plot shows the seasonal dependency of the Dobson No. 104 but it shows also the comparable temporal course of the Brewers and BTS Solar which is expressed by the least-square fit deviation of less than ±1% over the whole campaign.

Figure 6 shows the TOC slant path deviation (slant path = TOC·AMF) of the Brewer No. 010 (single monochromator) and BTS Solar compared to the Brewer No. 226 (double monochromator). This figure is a measure of the TOC evaluation with respect to air mass factor (AMF). AMF is dependent on the solar zenith angle (SZA) due to the longer attenuation path through the atmosphere at high SZA and the deviations should ideally be independent on this factor. As illustrated the BTS Solar is less dependent on the slant path as the Brewer No. 010 (slope -0.00174 %/DU·AMF) but a small dependency (slope -0.00056 %/DU·AMF) is recognizable compared to Brewer No. 226.
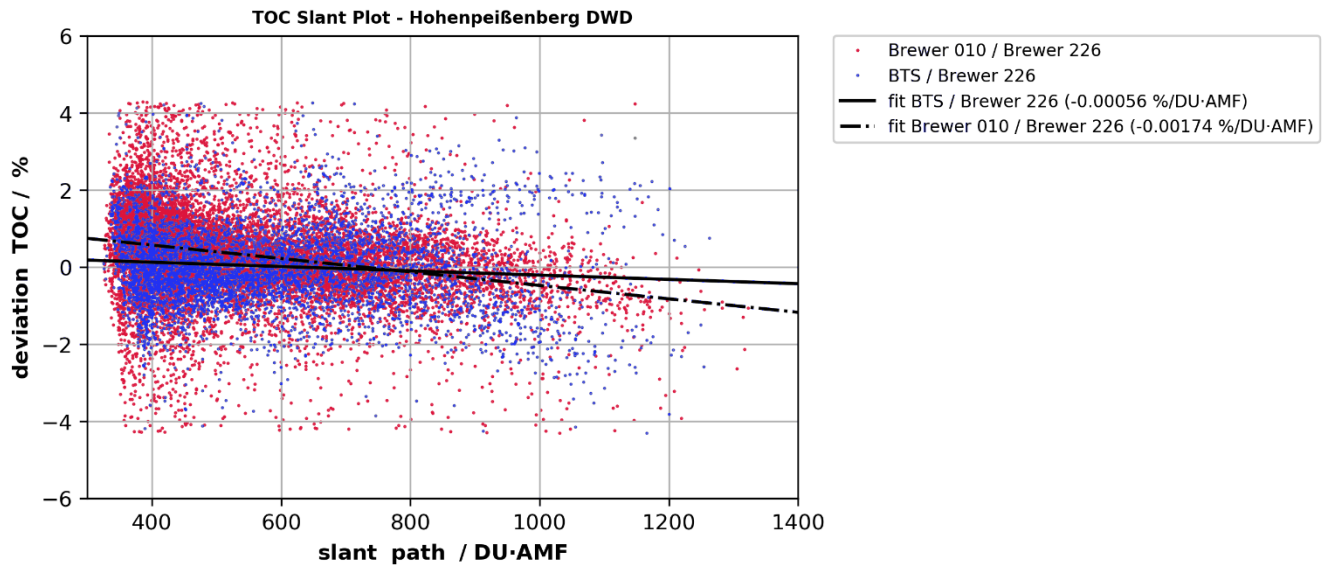
Figure 6 The TOC Slant plot for BTS Solar of all data shows a smaller air mass factor (AMF) dependency than the single monochromator Brewer No. 010 for the BTS Solar. Linear fits are used.

3.2 Koherent

Figure 7 presents exemplary daily courses of TOC from Koherent and Brewer 163 during the winter day of 2nd of February 2020 and the summer day of 27th July 2020, respectively.
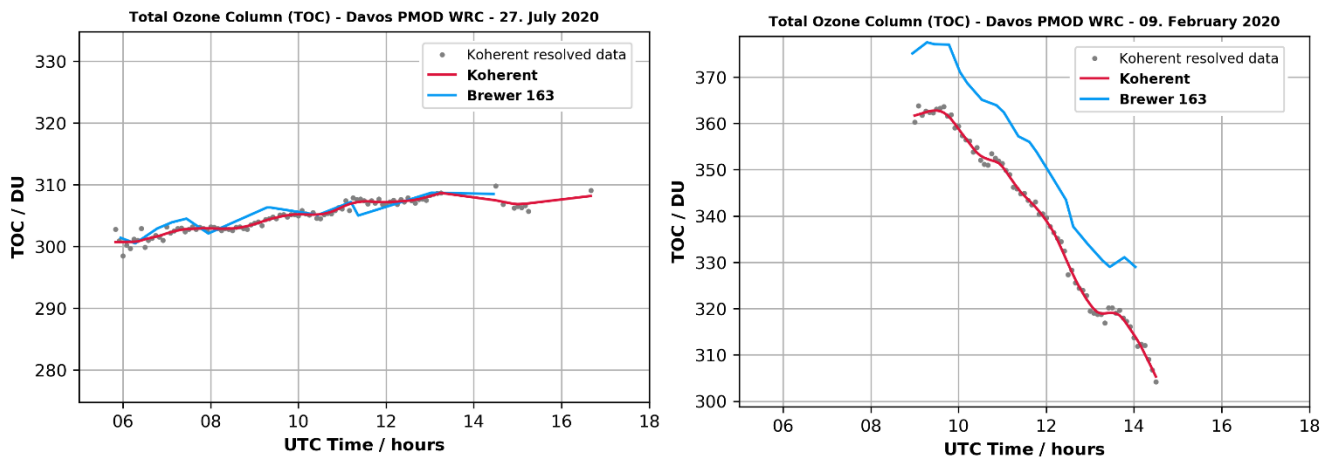
Figure 7 Two exemplarily days of Koherent vs. Brewer No. 163 in winter and summer season. These days illustrate the high temporal resolution of Koherent and show the seasonal dependency of the deviation.

One can see that the daily variation is well reproduced by Koherent with a high temporal resolution and its temporal development is in line with the values of Brewer No. 163. However, there is an absolute bias of TOC of around -10 DU compared to Brewer 163 depending on the season. In winter season Koherent shows a larger offset than in summer season, where the values are almost congruent.

Figure 8 shows the temporal course of the relative difference of TOC values between Koherent and Brewer No. 163 during the entire comparison period between 1st of October 2019 and 30th of December 2020 ((Koherent-Brewer163)/Brewer163). The least-square fit in the figure reveals clearly the seasonal dependency of up to 4% of the differences through the seasons. The differences are about -3% in summer season and about +1% in winter season in respect of the fit in figure 8.

Figure 9 presents the histogram of all synchronous data of the difference of Koherent and the Brewer No. 163. The histogram of the percent differences of TOC reveal a systematic overestimation by Koherent of an average of 1.64% during the entire period, with an expanded standard deviation of about 2.7% (*k=2*). This variation reflects mainly the seasonal s variablity and less the measurement error of the system, which is assumed to be comparable to the BTS Solar system (see section 3.1).

In order to compare the slant path dependency of the instrument, Figure 10 demonstrates the deviation of the differences as a function of the ozone slant path, defined as the multiplication of the AMF and TOC. Figure 10 a) shows a strong ozone slant path dependency with a slope of -0.0033%/TOC·AMF that is observed with Koherent. For further comparison, the slant path dependency is determined for the summer month of July 2020 only (Figure 10 b)), with air masses between 1.1 and 3.4 and stratospheric temperatures ranging from 230.2 K to 231.1 K. The slope of the linear fit of -0.0015%/TOC·AMF indicates that the slant path dependency is significantly lower than for the entire period. Figure 10 b) also include a polynomial fit of degree=2 (quadratic), highlighting that for slant paths below 800 DU the dependency shows almost no trend. For higher slant paths, however, the differences are increasing.
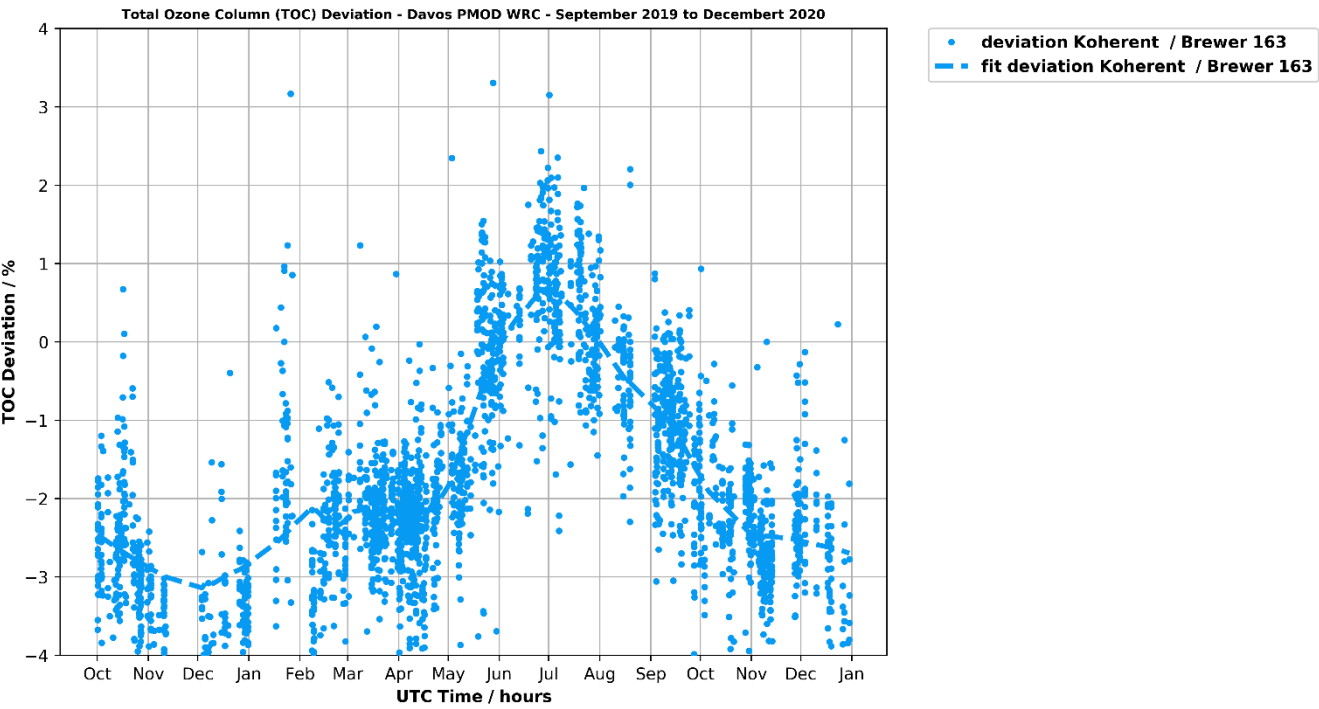
Figure 8 Temporal course of the relative differences of Koherent and BrewerNo. 163 during the entire period of comparison. The dashed line shows a least-square fit of the data.
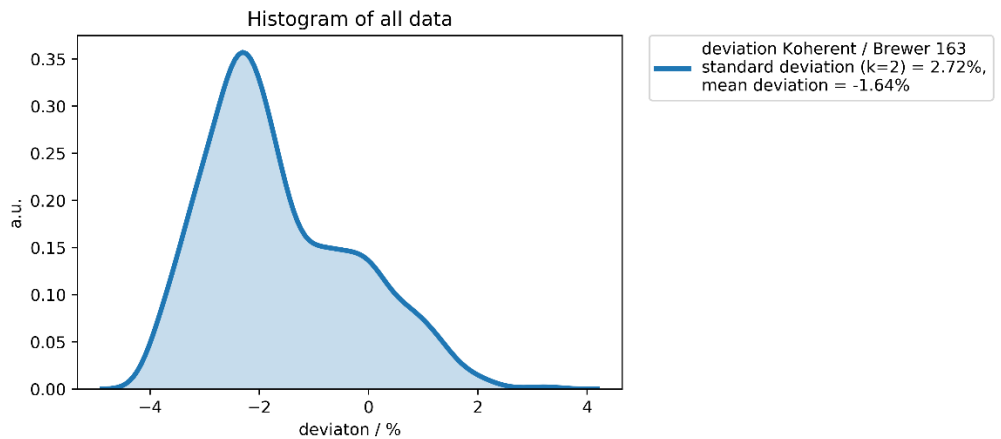
Figure 9 Histogram of the relative differences of Koherent and Brewer No. 163. The mean difference between both instruments is -1.64% revealing a clear bias of TOC. The standard deviation (*k=2*) of 2.72%, reflects the seasonal variation of the deviation.
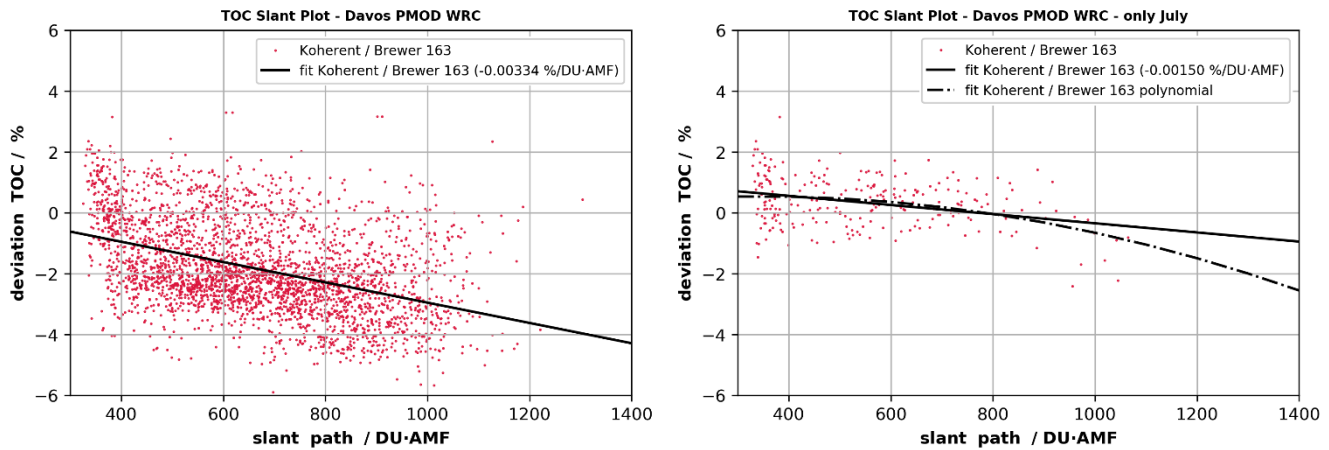
Figure 10 a) and b). Slant path dependency of Koherent vs. Brewer No. 163 a) for the entire period of comparison and b) for the month of July 2020 only including a quadratic polynomial fit additional to the linear fits.

4 Discussion

In this longer-term intercomparison of more than one year the array-spectroradiometer based BTS systems demonstrated their instrument performance published in Zuber et al. (2018b) and Egli and Gröbner (2018) for TOC determination as well during harsh environmental conditions like in Davos or Hohenpeißenberg. Both BTS setups, the fibre coupled entrance optic version by PMOD/WRC called Koherent and a diffusor optic based version by Gigahertz-Optik GmbH called BTS Solar where running the whole campaign without any significant technical issues.

The BTS Solar demonstrated to be robust and accurate in terms of stability since no change in responsivity was observed during the whole measurement campaign period (differences below the uncertainty of the calibration). The used TOC retrieval algorithm, based on a Look-Up-Table (LUT), demonstrated as well to be robust and accurate in terms of TOC determination over all seasons. Apart from the current air pressure (corrections of <0.8 DU over the full measurement campaign) no further parameters such as ozone temperature or other measured parameters are required as input of the algorithm for the evaluation at Hohenpeißenberg. Only standard parameters of the measurement location such as height and position are needed. Based on these parameters two (summer and winter) LUTs are calculated for a specific station with the libRadtran software package and later on just an air pressure correction is applied. This seems remarkable, since already by this simple modelling accurate TOC results over the whole season have been achieved at this site.

The retrieval algorithm of BTS solar is principally a partial spectral fit of two wavelengths bands, which substantially differs from the four wavelengths retrieval from Brewers and double wavelengths pairs from Dobsons or the least-square fit retrieval of Koherent. The partial spectral fit technique with the usage of a LUT demonstrated its performance so far in very different places such as Izaña Atmospheric Observatory in Tenerife (Zuber et al., 2018b), Huelva (not scientifically published yet) and now in Hohenpeißenberg. The algorithm allows worldwide usage (due to different available atmosphere models of libRadtran) but may need a finer parametrization at other locations worldwide or for special atmospheric profile situations which may not be tested yet. For the slant path dependency, a slope of -0.00056 %/DU·AMF was achieved, which is significantly lower than of single monochromator Brewer (-0.00174 %/TOC·AMF) compared to double monochromator Brewers.

Contrary to BTS solar, the TOC retrieval from Koherent is using the full spectrum retrieval in the spectral range of 305 nm and 350 nm. The minimal least-square algorithm as of Huber et al. (1995) and Vaskuri et al. (2018) includes for each TOC retrieved data point also the fitting of the aerosol optical depth This procedure of including the aerosol optical depth as a free fitting parameter of the algorithm allows the operational use at other locations worldwide with different atmospheric conditions.

Currently, the stratospheric temperature is not retrieved by the algorithm and a climatological averaged value of -45°C is chosen instead. . The strong seasonal trend correlates with the variation of the stratospheric temperature at Davos. Effective stratospheric ozone temperatures from ozone launches in Payerne (Switzerland) or from the ECMWF reanalysis as presented in Gröbner et al. (in review 2021) show a difference of about 10 K between summer season and winter season correlating with the differences in TOC. This indicates that the algorithm for Koherent may be strongly sensitive to stratospheric temperature. In a future version of the algorithm we will investigate if stratospheric temperature can be retrieved from the measurements or be included in the retrieval.

The strong ozone slant path dependency of Koherent in Figure 10 also reflects this seasonal trend of the deviations, since larger SZA and corresponding higher air-masses are more pronounced in winter season than in summer season. Figure 10 b) shows the ozone slant path dependency restricted to one month with an almost constant stratospheric ozone temperature of around 230 K. The graph reveals, that the slant path dependency is significantly lower (-0.0015%/TOC·AMF) than for the entire period over all seasons (-0.0033%/TOC·AMF)- This supports the conclusion that the strong slant path dependency is caused by biases due to stratospheric temperature differences.

However, slant path dependency could also be caused by stray light, which biases the ozone at large airmasses. As mentioned in the Introduction and stated in (Zuber et al., 2018a), the BTS array-spectroradiometer suffer from straylight at short wavelengths (below 310 nm) at high SZA. A potential effect of this remaining straylight can explain the increase of the differences at high slant paths above 900 DU as shown in Figure 10 b). The slope of the slant path dependency of -0.0015%/TOC·AMF is comparable to the slant path dependency between a single and a double monochromator Brewer with a slope of -0.0017%/TOC·AMF (Figure 6). However, a proper assessment of the effect of straylight from Koherent, can only be addressed when the effect of stratospheric temperature is corrected. Based on this dataset, where the stratospheric temperature is not included, the biases from stratospheric ozone temperature seems to be much stronger than a potential effect of straylight.

For BTS Solar, potential straylight impact on TOC is less pronounced. In figure 2, a slight deviation of 5 DU can be observed in the early morning at high SZA. The statistical analysis of the slant path dependency reveals a linear slope of -0.00056%/TOC·AMF, which is less than for single monochromator Brewers. Practically the measurement range of TOC is comparable since also the TOC values of double monochromator Brewer at high SZA/AMF are usually not used due to too low SNR (e.g. DWD applies an AMF filter >3.5 single monochromator Brewer or AMF >4 double monochromator to TOC values. These values were determined on the basis of long-term experience.). We assume that the impact of straylight is less pronounced than for Koherent, because the first wavelength band of the BTS Solar is an average between 307 nm and 311 nm, where less straylight can be expected, than using the full spectrum until 305 nm for the least-square fit algorithm. Most likely the least-square fit algorithm of Koherent is more sensitive to biases at short wavelengths.

The calibration.of Koherent showed to be stable over to whole measurement campaign, however an offset was detectable.-We assume that the large variation of 2.2% (k=1) from the instrument responsivities is based on the calibration difficulty of a lens-based telescope for an irradiance calibration. The alignment of the telescope-based system seems to be very sensitive on distances between Lamp and the collimating lenses and the angular orientation of the telescope.

In the future, a recalibration of Koherent is planned with an improved calibration setup and comparison with additional reference instruments.

In this respect the irradiance calibration of the BTS Solar seems to be easier applied, since only a global diffusor system has to be calibrated using standard irradiance calibration with easily accessible reference plane instead of a lens-based telescope. The fibre-coupled telescope system has a significantly lower light throughput and is therefore more sensitive to noise at short wavelengths.

In general, the BTS Solar is comparable to the Brewer No. 010 with a mean deviation of 0.04% and an expanded standard deviation ($k=2$) of 1.29% and Brewer No. 226 with a mean deviation of 0.06% and an expanded standard deviation ($k=2$) of 1.47%. The Dobson No. 104 showed a mean deviation of -0.84% and an expanded standard deviation ($k=2$) of 2.22%. Koherent achieved a deviation of 1.64% with an expanded standard deviation ($k=2$) of 2.72% by showing a significant seasonal peak of up to 4%.

In the discussion of replacing standard ozone observing instruments, the comparison of Brewers and Dobsons has a long tradition. It was observed that using the Paur and Bass (1985) cross-section, the Brewers and Dobsons show a seasonal difference depending on the stratospheric temperature. Redondas et al. (2014) showed that this difference is significantly smaller when using the IUP cross-section (Serdyuchenko et al., 2013). Recently, Gröbner J. (2021) compared Brewers and Dobsons using other different cross-sections and including stratospheric temperature. The study, especially the data of Koherent, highlights again the importance of applying a specific cross-section and its stratospheric temperature dependency in order to homogenize TOC data from different co-located instruments. From this point of view the new systems in general should be further compared at different locations with different atmospheric conditions and the optimal cross-section for the best homogenization of the data should be systematically investigated.

5 Conclusion and Outlook

In summary, the BTS Solar performance regarding TOC determination with data of more than one year is comparable to the performance of the Brewers used in this study. The slant path dependencies (slope of the curve) of the BTS Solar may hint to some remaining straylight at high SZA (above 65°), which is less than a single monochromator Brewer but larger than of a double monochromator Brewer.

The LUT retrieval algorithm used at Hohenpeißenberg was insensitive to stratospheric temperature and provided accurate results by a simple modelling. However, a validation in the presence of significant SO_2 or aerosol levels could not be tested. The performance in specific other stations regarding the atmospheric composition should be further investigated.

Koherent showed a comparable performance to a double monochromator Brewer regarding TOC determination in summer time (around 1% bias), however, with a significant seasonal drift in the winter time of down to -3%. The least-square fit algorithm of Koherent is expected to be insensitive to atmospheric changes, due to the included fitting of aerosol optical depth. Currently, the algorithm of Koherent is sensitive to stratospheric temperature, which explains the seasonal variability relative to the Brewer No. 163. In a future version of the retrieval algorithm, the stratospheric temperature will either be retrieved by the algorithm, or added as an additional parameter to the retrieval routine in order to reduce the observed seasonal variation.

Data availability. The datasets used in this study can be downloaded from https:// doi.org/10.6084/m9.figshare.14686656

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

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