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Total column ozone measurements by the Dobson spectrophotometer at Belsk (Poland) for the period 1963–2019: homogenization and adjustment to the Brewer spectrophotometer

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Abstract. The total column ozone (TCO₃) measurements by the Dobson spectrophotometer (serial no. 84) have been carried out at Belsk station (51°50′ N, 20°47′ E), Poland, since 23 March 1963. In total, ~ 115000 intraday manual observations were made by 31 December 2019. These observations were performed for different combinations of double wavelength pairs in the ultraviolet range and observation types, i.e., direct sun (DS), zenith blue (ZB), and zenith cloudy (ZC) depending on weather conditions. The long-term stability of the instrument was supported by frequent (almost every 4 years) intercomparisons with the world standard spectrophotometer. Trend analyses, based on the monthly and yearly averaged TCO₃, can be carried out without any additional corrections to the intraday values. To adjust these data to the Brewer spectrophotometer observations under low solar elevation, presence of clouds, and the temperature dependence of ozone absorption. The adjusted time series shows that the Brewer–Dobson monthly averaged differences are in the range of about ±0.5%. The intra-day TCO₃ database, divided into three periods (1963–1979, 1980–1999, and 2000–2019), is freely available at https://doi.org/10.1594/PANGAEA.919378 (Rajewska-Więch et al., 2020).

1 Introduction

The monitoring of total column ozone (TCO_3) started in 1924 in Oxford (the United Kingdom) with a prototype of the Dobson instrument (DI). Before the Second World War, there existed data records from two stations – Oxford (DI serial no. 1) and Arosa (DI serial no. 2) – archived in the World Ozone and Ultraviolet Radiation Data Centre. After the international geophysical year in 1958, the total number of the stations with routine TCO₃ increased up to about 50. The number of ozone observations increased sharply in the early 1980s following the discovery of the ozone hole in the Antarctic spring by Chubachi (1984) and Farman et al. (1985). The variability in the ozone layer was of particular interest as ozone is the only gaseous component of the atmosphere that absorbs short-wave solar radiation in the ultraviolet (UV) B range (280–315 nm), which is harmful to all living organisms on Earth. Ozone depletion became a widely debated environmental issue that led to the signing of an international agreement, the Montreal Protocol (MP) 1987, to protect the ozone layer by reducing the production and use of the ozone-depleting substances (ODSs).

Until the late 1970s the Dobson spectrophotometer was the only optical instrument for TCO_3 ground-based measurements. Monitoring of TCO_3 required a lot of man power because the Dobson observations involve many manual activities, and the obtained TCO_3 value depended to some extent on the individual observer's ability to manipulate the instrument. The need to automate TCO_3 measurements resulted in the development of an automated instrument – the Brewer spectrophotometer. It was designed in Canada, and after improvements in the 1970s, it was available for ozone monitoring in the early 1980s. However, differences between the Dobson and the Brewer record data were found. Kerr et al. (1988) suggested that this difference was due to the temperature dependence of the ozone absorption coefficients (O₃AC). The Bass–Paur O₃AC (Bass and Paur, 1985), which replaced the previous Vigroux (1967) coefficients, has been used operationally in the TCO₃ ground-based observation network since the early 1990s (Komhyr et al., 1993). Vanicek (2006) found that the difference could reach ~ 4 % when comparing the Brewer and Dobson monthly mean data taken in Hradec Králové, Czech Republic, due to the use of the Bass–Paur O₃AC at a fixed temperature. There were several attempts to recalculate the O₃AC (Redondas et.al., 2014), and finally the recalculation proposed by Serdyuchenko et al. (2014) was recommended for the ground-based TCO₃ network. The application of these temperature-dependent absorption coefficients significantly reduced the artificial seasonality in the Dobson-Brewer differences to less than 1 % (Redondas et al., 2014; Fragkos et al., 2015). However, the data from the Dobson network have not yet been recalculated using the new O₃AC as the vertical temperature and ozone profiles in the stratosphere are also required for the observation site.

The ozone issue is still relevant as the rate of ozone recovery expected from the regulations of the MP 1987 and its subsequent amendments was also driven by recent climate changes (e.g., Steinbrecht et al., 2017; Ball et al., 2018). In addition, a surprising increase in the concentrations of anthropogenic chlorofluorocarbons in the troposphere has been found in some regions related to ODS leakage from its long-term storing reservoirs (Lickley et al., 2020) and a return to industrial use of ODSs (Montzka et al., 2018; Dhomse et al., 2019). The Antarctic ozone hole is surprising because of its variability from an extreme small extent in 2019 (Krzyścin, 2020) to extreme large one in 2020 (https://public.wmo.int/en/media/ news/2020-antarctic-ozone-hole-large-and-deep?, last access: 2 September 2021). Moreover, severe chemical losses occurred in the Arctic stratosphere in spring 2020 (e.g., Manney et al., 2020; Wohltmann et al., 2020). Therefore, it is still worth monitoring ozone with the Dobson spectrophotometer, which was designed and put into operation almost 100 years ago.

The Central Geophysical Laboratory of the Institute of Geophysics, Polish Academy of Sciences, at Belsk $(51.84^{\circ} \text{ N}, 20.78^{\circ} \text{ E})$, Poland, started monitoring atmospheric ozone (TCO₃ and the ozone vertical profile by the Umkehr method) on 23 March 1963. There are only two stations in Europe with longer time series: Arosa (since 1926; Staehelin et al., 1998) and Hradec Králové (since 1961; Vaníček et al., 2012). The importance of Belsk's TCO₃ time series results from the monitoring carried out regardless of the weather conditions, excluding only days with continuous rain or snow fall. Various efforts have been made to support data quality (e.g., Dziewulska-Łosiowa et al., 1983; Degórska and Rajewska-Więch, 1991) because less accurate zenith sky TCO₃ observations during cloud cover were only available on certain days, especially in late autumn and winter. This paper presents further steps in the homogenization of Belsk's TCO₃ time series for the period 1963–2019 in the perspective of the long-term variability in the atmospheric ozone and possibility of replacing Belsk's Dobson with a modern Brewer spectrophotometer.

2 Data and methods

2.1 The Dobson spectrophotometer

The Dobson spectrophotometer was designed to measure TCO₃ by the technique of differential optical absorption spectroscopy (Dobson, 1957). The TCO₃ values from direct sun (DS) observations are calculated based on the Beer's law applied to spectral UV solar irradiances for selected wavelengths with strong and weak absorption by the ozone layer. These wavelengths are denoted as A (305.5–325.0 nm), C (311.5–332.4 nm), and D (317.5–339.9 nm). The TCO₃ value is determined from linear combination of the logarithm of the ratios between A pair and D pair irradiances, called the double AD wavelength pair algorithm, to reduce the effects of the aerosols scattering on calculated ozone. It is also possible to use C pair irradiances instead of the A pair, i.e., the so-called double CD pair, to obtain TCO₃ if the solar irradiance at the shorter wavelength of the A pair is too weak because of a large ozone absorption under high air masses $(\mu > 3)$ and/or during cloudy conditions.

Another option to calculate TCO₃ by the Dobson spectrophotometer is to use scattered sunlight from the zenith sky (ZS) at the same wavelengths as for the DS observations. In this case, the same algorithm is implemented, but the result needs to be reduced by empirical charts. The charts proposed by Rindert (1973) were used throughout Belsk's observations for AD and CD double wavelength pairs. The operational data archived in the PANGAEA database was obtained using the Bass–Paur O₃AC at fixed temperature of -46.3 °C according the recommendation of Komhyr et al. (1993).

Figure 1 shows time series of the number of daily TCO₃ means obtained from DS intraday observations with AD and/or CD wavelength pairs in January and July for the 1964–2019 period. The number of the DS daily means per month is larger in June because of usually less cloudy conditions this month compared to January and longer day length in summer (i.e., greater chance for cloudless conditions). The monthly mean TCO₃ for January is mostly based on ZS observations. It is worth mentioning that a reduced number of DS observations (less than 15 per month) were found in July for the period 1985–1995. During this period, ZS observations under a cloudless sky, i.e., the so-called zenith blue (ZB) observations, replaced DS observations because the so-lar disk was not clear due to heavy contamination of Belsk's



Figure 1. Number of TCO_3 daily means based on the direct sun measurements for January and July in the period 1964–2019.

atmosphere with industrial aerosols. The observers were then recommended to choose ZB measurement for a clear sky.

The TCO₃ values by ZS observations could be calculated for almost all weather conditions (excluding cases with heavy and variable cloudiness), but they are reliable, i.e., close to the DS values, for the air masses up to 2.8 (Fig. 2a). The empirical reduction curves (Rindert, 1973), which were used to convert ZS values to equivalent DS values, did not reflect all possible combinations of cloud types and their positions in the sky. Therefore, the DS–ZS differences can sometimes exceed 5 %. To eliminate the long-term drift of the DS– ZS differences, the following transfer function is used from the regression line fit to the relative differences between DS and ZS TCO₃ subsets for $\mu \subset [2.8, 4.0]$:

$$TCO_{3, ZS}^{*} = (1 + 0.0125(\mu - 2.8)) \times TCO_{3, ZS},$$

for $\mu \subset [2.8, 4.0],$
$$TCO_{3, ZS}^{*} = 1.015 \times TCO_{3, ZS}, \text{ for } \mu > 4.0,$$
 (1)

where TCO_{3, ZS} is the TCO₃ value derived from ZS observations, μ is air mass during the Dobson observation, and TCO_{3, ZS}^{*} pertains to the corrected value for $\mu > 2.8$. However, the linear correction is not valid if $\mu > 4.0$ (Fig. 2a). Only 0.7 % of all TCO₃ observations were made at such high μ values. The fixed correction of 1.015 is applied if $\mu > 4.0$, but TCO_{3, ZS}^{*} values should be treated with caution. The smoothed pattern of the DS–ZS differences (Fig. 2b) is close to zero after the application of the transfer function according to Eq. (1).

DS–AD observations provide the most accurate TCO₃ values (Dobson, 1957) when $\mu < 2.5$. However, these observations are not possible for northern sites, such as Belsk, where the noon air masses above 2.5 appear in the period from 3 November up to 9 February, and the yearly maximum air mass at noon on 23 December is 3.75. Therefore, DS–CD observations are recommended for these periods (Dobson, 1957). Additional intraday TCO₃ values, especially af-

ter 2005, were calculated using the nearly simultaneous DS– AD and DS–CD observations to find a relationship between these TCO₃ values. Figure 3a shows that the relationship is quite stable in the 1963–2019 period, i.e., the smoothed differences (black curve) are within the [0%, 1%] range, and the linear regression line (blue line) is trendless. A similar comparison between DS and ZS observations, after application of the transfer function (1), ensures that the smoothed differences are close to the zero line throughout the observation period (Fig. 3b).

The monthly mean TCO₃ differences between various subsets of the TCO₃ data over the period 1963–2019 are shown in Fig. 4. The differences are derived from the monthly averaging of the daily mean TCO₃ values for the following subsets: DS-AD, DS-CD, ZB, ZS, and for less reliable ZS observations, when the zenith was obscured by clouds, i.e., the so-called zenith cloudy (ZC) observations. For all ZS intraday values if $\mu > 2.8$, the correction function according to Eq. (1) was applied. The TCO₃ differences shown in Fig. 4 are in the range of ± 1 % almost all year round, excluding autumn/early winter months (October-November-December). Moreover, for each selected month, from January to September, the spread between the monthly mean differences is about ~ 1 % for the all considered subsets of the Dobson observations. The maximum spread is 4 % in December based on the difference of -1% for DS-AD vs. DS-CD and 3% for DS vs. ZC.

2.2 Calibrations of the Dobson spectrophotometer

The Dobson spectrophotometer serial no. 84 at Belsk has been operated continuously since 23 March 1963 in the same place with only a few breaks in observations (up to several weeks) due to the international calibration campaigns with other Dobson instruments. The frequent comparisons of Belsk's Dobson with the world standard instrument in combination with the recalibration of the optical wedge of the instrument and the calculation of the resulting R–N tables were of key importance for maintaining the quality of Belsk's Dobson. The R–N table is used to convert the dial reading (the so-called R value) obtained by the Dobson observer into the logarithm of the ratio between the light intensities in a pair of the UV wavelengths with weak and strong absorption by ozone (the so-called N value). N values are used in theoretical formulas to calculate TCO₃ (e.g., Dobson, 1957).

The first DI intercomparison was held at Belsk in June/July 1974. The participating Dobsons were compared against the world primary standard (WPS) Dobson spectrophotometer no. 83 and the optical wedge of Belsk's Dobson was recalibrated (Dziewulska-Łosiowa and Walshaw, 1975). The next DI intercomparison was held in Potsdam (Germany) in June 1979 also with the attendance of the same WPS. During the comparison the new R–N tables were obtained, which only slightly differed from those calculated earlier at Belsk's intercomparison (Dziewulska-Łosiowa et



Figure 2. Air mass dependence of differences between daily mean TCO_3 values by DS and ZS measurements as a percentage of the DS TCO_3 values: original data (a) and ZS TCO_3 after the correction using Eq. (1) (b).



Figure 3. Time series of relative differences between daily means of TCO_3 : DS–AD vs. DS–CD measurements (a) and DS vs. ZS measurements (b). The black curve shows the smoothed pattern of the data. The blue line is the linear regression line.



Figure 4. The monthly mean relative differences between daily TCO₃ means derived from different types of the Dobson measurements. The bar denotes the range: mean \pm standard deviation.

al., 1983). The subsequent campaigns were in Arosa (1986, 1990, and 1995) with the WPS Dobson no. 83. The next intercalibrations (2001, 2005, 2009, and 2014) were organized at the European regional Dobson calibration center, the Me-

teorological Observatory Hohenpeissenberg, Germany. The local European sub-standard, DI no. 64, was used as the reference instrument for comparisons at the center. After the intercomparisons, the new R–N tables were obtained and used immediately, i.e., after the instrument arrived at the observing site, as input to the TCO₃ calculation algorithm. The retrieval software has not been changed since its first operational use in 1978 (Degórska et al., 1978). Table 1 summarizes the intercomparison campaigns with Belsk's Dobson.

The first homogenization of Belsk's TCO₃ data for the period 1963–1981 by Dziewulska-Łosiowa et al. (1983) was based on Potsdam's R–N tables, extraterrestrial constants determined at Belsk in 1974 during the first DI intercomparison, and the O₃AC by Vigroux (1967). The zenith sky irradiances (for AD and CD observations) were reduced to the DS equivalent values using the empirical charts according to Rindert (1973). The next homogenization was in 1991 by Degórska and Rajewska-Więch (1991). All data collected before 1992 were re-evaluated on a reading-by-reading basis using the Bass–Paur O₃AC (at a fixed temperature of -46.3 °C) that has been recommended for processing Dobson TCO₃ measurements since 1 January 1992.

Optical wedge calibrations performed during the entire period of ozone observations at Belsk had only a slight effect

Site/country	Year	Standard instrument
Belsk/Poland	1974	World standard Dobson no. 83
Potsdam/Germany	1979	World standard Dobson no. 83
Arosa/Switzerland	1986	World standard Dobson no. 83
Arosa/Switzerland	1990	World standard Dobson no. 83
Arosa/Switzerland	1995	World standard Dobson no. 83
Hohenpeissenberg/Germany	2001	European sub-standard Dobson no. 64
Hohenpeissenberg/Germany	2005	European sub-standard Dobson no. 64
Hohenpeissenberg/Germany	2009	European sub-standard Dobson no. 64
Hohenpeissenberg/Germany	2014	European sub-standard Dobson no. 64

 Table 1. The intercomparison campaigns with the Dobson instrument from Belsk.

on the TCO₃ values. Figure 5 shows the comparison of TCO₃ calculated for the first 2-year period (1963-1964) and last 2year period (1980-1981) of the time series after the first homogenization in 1983. Original TCO3 values from DS-AD observations presented by Dziewulska-Łosiowa et al. (1983), which were based on the Vigroux O₃AC, are converted to the values obtained using the Bass-Paur O3AC (i.e., multiplied by 0.9743, as recommended by the International Ozone Commission in 1992) and then compared with the corresponding values obtained from the current retrieval based on the Bass-Paur O₃AC and R-N tables obtained in the last intercomparison in 2014. There is almost one-to-one correspondence between the two TCO₃ datasets, i.e., the slope of linear fit is 0.9999 and 0.9962 for the periods 1963–1964 and 1980–1981, respectively. The maximum difference between TCO₃ values is less than 1 %. Small differences (< 1 %) are also found after comparison of the DS-AD subset of TCO3 values in August 2020 calculated using different R-N tables that were operationally used in the 1990s and 2020s (Fig. 6).

From these comparisons of the DS–AD measurements, it can therefore be concluded that Belsk's Dobson demonstrated stable performance throughout the measurement period. A linear approximation of TCO₃ values in periods between the calibration campaigns was not necessary to account for changes in R–N values and instrument aging.

2.3 Adjustment to the Brewer spectrophotometer

The ozone monitoring with the Brewer spectrophotometer serial no. 64 (BS64) mark II (single monochromator) was launched at Belsk in 1991. Like other Brewers, the BS64 is a fully automated, self-testing, PC-controlled instrument intended for continuous, long-term observations in all weather conditions (e.g., Fioletov et al., 2005). The quality of BS64 measurements has been supported by regular (yearly or at least every 2 years) comparisons with the international travel reference instrument provided by the International Ozone Services Inc. (https://www.io3.ca/index.php, last access: 2 September 2021). The BS64 instrument constants re-defined after each comparison were immediately incorporated into the operating retrieval.



Figure 5. TCO₃ daily means by AD–DS measurements in the periods 1963–1964 and 1980–1981 calculated with the present (2019) calculation algorithm (R–N tables based on the 2014 intercomparison) vs. those by the same algorithm using old (1979) R–N tables.

The Brewer TCO₃ is derived from a weighted linear combination of solar irradiances at five wavelengths (306.3, 310.0, 313.5, 316.8, and 320.0 nm) to eliminate noise due to SO₂ absorption in the UV range (Kerr et al., 1988). The Brewer TCO₃ retrieval uses the Bass–Paur O₃AC at -45 °C for two types (DS and ZS) of observations. There are many daily BS observations regardless of weather conditions. Cycles of five observations, which are performed in \sim 3–4 min, are repeated every 5 to 20 min throughout the day depending on the instrument's schedule. The mean TCO3 value, averaged over five observations, is calculated if the scatter of these TCO₃ values is small, i.e., standard deviation is less than 2.5 Dobson units (DU). The Dobson spectrophotometer provides instantaneous TCO₃ values, while the Brewer instrument gives the average of five observations, so this may be an additional source of differences between the spectrophotometers.

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Figure 6. Daily means of TCO₃ in August 2020 from DS-AD measurements as calculated with different R-N tables obtained during intercomparisons in the 1990s and 2000s (see Table 1).

231 Correction for the effective temperature

Figure 7a shows the time series of the differences between Brewer and Dobson TCO3 daily means (Brewer minus Dobson TCO₃ as percent of the Dobson TCO₃ value) calculated for the period 2002-2019. This period was selected because more DS-AD and DS-CD observations were performed during the day (see Fig. 3a) to construct a transfer function from the Dobson TCO_3 to the Brewer equivalent TCO_3 before the predicted change in monitoring policy to less frequent measurements during the day, i.e., only a few observations around noon in perfect weather conditions. The well-known seasonality of the Brewer-Dobson (BD) differences, i.e., underestimation of the Dobson TCO₃ in the cold period of the year, could be observed after applying a low-pass filter, the locally weighted scatterplot smoothing (LOWESS, Cleveland, 1979), to the time series of BD differences.

To reduce the BD seasonal differences, the temperaturedependent O₃AC is applied instead of the operational O₃AC assuming fixed temperature T_0 , i.e., $T_0 = -46.3 \,^{\circ}\text{C}$ (Dobson) and $T_0 = -45.0$ °C (Brewer). Here we use the temperature correction factor, α , calculated by Redondas et al. (2014), i.e., $\alpha = 0.104 \% \text{ K}^{-1}$ (for Dobson) and $\alpha = 0.009 \% \text{ K}^{-1}$ (for Brewer). These values are based on the O₃AC dependence on temperature obtained by the Institute of Experimental Physics (IUP), University of Bremen (Serdyuchenko et al., 2014). The correction function to account for the actual effective temperature is as follows:

$$TCO_{3, NEW} = (1 + \alpha (T_0 - T_{eff})) \times TCO_{3, OLD}, \qquad (2)$$

where $T_{\rm eff}$ is the effective (weighted by the ozone vertical profile) temperature, TCO_{3. OLD} is the TCO₃ value at the fixed temperature, and TCO3, NEW is the temperaturecorrected value. Prior to the application of Eq. (2), TCO₃ values by ZS Dobson observations need to be converted to the DS TCO₃ equivalent using transfer function according to Eq. (1). The effective temperature at noon is taken after the Tropospheric Emission Monitoring Internet Service (TEMIS) overpass data (https://www.temis.nl/climate/ efftemp/overpass.php, last access: 2 September 2021), based on the European Centre for Medium-Range Weather Forecasts (ECMWF) model estimates. Koukouli et al. (2016) found that the ECMWF effective temperature was in good agreement with that from the ozone sounding in Northern Hemisphere midlatitudes.

Figure 7b shows the time series of the BD differences after applying the correction function according to Eq. (2). The visible seasonality in Fig.7a disappears, but there are still limited seasonal variations. To find out the range of these oscillations, the monthly mean BD differences are calculated averaging the daily differences. The results are shown in Fig. 8a (DS subset) and Fig. 8b (ZS subset). A comparison of the original DS and ZS data (not accounting for $T_{\rm eff}$ seasonal variability) ensures that the seasonal BD difference is ~ 4.5 % as calculated using the monthly mean difference values of ~ 3.5 % and -1 % for January and July (or August for ZS data), respectively. After the application of Eq. (2), the BD difference is smaller, i.e., $\sim 2.5\%$ (DS) and 3%(ZS). The smoothed curve in Fig. 7b ensures that the Dobson TCO₃ values were ~ 1 % lower (2002–2004) and ~ 1 % higher (2018-2019) compared to the Brewer values. Such discrepancies may be related to the Brewer ZS TCO₃ values because they may be influenced by clouds (the Brewer ZS algorithm is based on a statistical relationship with parallel DS observations), which in some years results in overestimation (or underestimation) in relation to the Dobson TCO₃ values.

It is worth mentioning that the monthly BD differences for the temperature-corrected data are out of the ± 1 % range only in January and December (DS subset) and additionally in November for the ZS subset. To the authors' knowledge, the BD differences for a less precise ZS subset have not yet been discussed by other authors. It seems that the stray light effect, which may be different in the Dobson and Brewer instruments (Belsk BS64 is a single monochromator spectrophotometer), is responsible for greater differences between these spectrophotometers during low-solar-elevation periods.

Correction for the stray light effect 2.3.2

The stray light within the Dobson instrument causes an underestimation of the TCO₃ value by about a few percent, when slant TCO₃, $\mu \times$ TCO₃, exceeds 800–900 DU (Evans et al., 2009). This underestimation also applies to the Brewer single monochromator spectrophotometers (e.g., Karppinen et al., 2015). Moeini et al. (2019) discussed the differences between TCO₃ values measured almost simultaneously by the Dobson and Brewer spectrophotometers due to the stray light effect. They found that the difference for low solar elevations (slant $TCO_3 > 800 DU$) was related to the instru-



Figure 7. The differences between the Brewer and Dobson TCO_3 daily means as a percentage of the Dobson TCO_3 value: original data after application of the correction function, according to Eq. (1), to the Dobson ZS data (a) and the Dobson and Brewer TCO_3 corrected for the effective temperature (b). The red curve represents the smoothed pattern by the LOWESS filter.



Figure 8. The monthly means of the Brewer–Dobson differences for the operational data (Bass–Paur O₃AC at fixed temperature) after application of the correction, according to Eq. (2), for the dependence of O_3AC on temperature based on IUP measurements and after adjustment of the Dobson TCO₃ to the Brewer TCO₃ equivalent by Eq. (3) accounting for the stray light effect differences between the spectrophotometers. The results are for DS measurements (**a**) and ZS measurements (**b**). Bar denotes the range: mean \pm standard deviation.

ment's individual sensitivity to stray light, which may be particularly high for a single monochromator Brewer (Brewer Mark II), i.e., the same type operating at Belsk.

Here, it is decided to construct a simple correction function, which would shift all-monthly BD differences for DS and ZS subset (Fig. 8) to the smallest possible range. The following the Brewer–Dobson adjustment (BDA) function is proposed to transform TCO_{3, NEW} (Dobson TCO₃ after the application of the transfer function according to Eq. 1 and the temperature correction according to Eq. 2) to its Brewer equivalent value denoted as TCO_{3, DOB→BRE}:

$$TCO_{3, DOB \to BRE} = (1 + \gamma \mu) \times TCO_{3, NEW}, \ \mu > \mu_0,$$

$$TCO_{3, DOB \to BRE} = TCO_{3, NEW}, \ \mu \le \mu_0,$$
 (3)

where constant values γ and μ_0 can be different for the ZS and DS subsets. The best pair of γ and μ_0 was experimentally derived examining many combinations of γ and μ_0 , taking values from the range of [0, 0.02] and [2.5, 3.5], respectively. Finally, the following constants are derived: $\gamma_{\text{DS}} = 0.5625 \times 10^{-2}$ and $\mu_{0, \text{ DS}} = 2.95$ (for DS subset), and

 $\gamma_{ZS} = 0.6250 \times 10^{-2}$ and $\mu_{0, ZS} = 2.8$ (for ZS subset). The corresponding ranges of the monthly mean BD differences are [-0.66 % (July), 0.46 % (January)] and [-0.48 % (December), 0.35 % (January)], for the DS and ZS, respectively. After application of the BDA function, the BD monthly mean differences are almost in the ± 0.5 % range (see Fig. 8 for the IUP and Brewer-adjusted subset).

2.4 Uncertainty of the Brewer-adjusted Dobson TCO₃

Typically, daily TCO₃ averages were archived based on a few measurements around local noon with nominal best quality. The AD–DS observation shows the highest accuracy of all possible combinations of the double wavelength pairs (AD and CD) and observation type (DS, ZB, and ZS). This kind of observation is not always possible because of weather conditions (clouds) and during low solar elevation. At the beginning of the TCO₃ observations at Belsk, a decision was made to increase the number of daily observations for selected days in each month in order to assess the uncertainty of TCO₃



Figure 9. The Brewer-adjusted TCO_3 values measured at Belsk on 8 August 2017 with different settings of the Dobson spectrophotometer.

observations. On such days, there were many, almost simultaneous observations with different instrument settings. For example, Fig. 9 shows the TCO₃ measurements at Belsk on 8 August 2017, planned to calculate differences between successive TCO₃ values. During the day, 46 observations were made for the following instrument settings: AD–DS, AD– ZS, and CD–DS.

The difference between two consecutive TCO_3 values sometimes exceeded 10 DU even if the measurements were taken within 10 min. It is impossible for TCO_3 to change rapidly over such a short timescale, taking into account natural variability in ozone in the stratosphere. The statistics of TCO_3 differences between the almost simultaneous Dobson measurement allows us therefore to estimate the uncertainty of the Brewer-adjusted Dobson TCO_3 values.

The statistics were obtained for various selected maximum ranges between the successive measurements, i.e. less than 1, 5, and 10 min. It was found that the statistical parameters did not depend on these ranges, which proved a reliable estimation of the uncertainty. This uncertainty combines the instrumental uncertainty associated with the differences between various types of Dobson measurements with the uncertainty resulting from the observer's skill to perform the measurements. Table 2 presents the statistics of the differences between TCO₃ values obtained almost simultaneously for different data subsets.

The uncertainty of the Dobson observations for the subset with the nominal highest accuracy (DS-AD) is [-1.15%, 1.09%], calculated as the range from the 5th to 95th percentile. The uncertainty increases with decreasing solar elevation, and the largest value, [-4.58%, 4.04%], is for the ZS subset when air mass is between 3 and 4. The greatest uncertainty for this type of observations seems to be due to the cloud variability over the site as the ozone retrieval for non-DS observations is based on a statistical relationship

Table 2. The statistics of the differences between the Breweradjusted Dobson TCO₃ values from two successive measurements taken at an interval of no longer than 10 min for the period 1963– 2019. The differences (in % of the mean of the two compared values) are shown for different sub-classes of the data and μ ranges. SD and *n* denote standard deviation and number of the measurement pairs used in the calculations, respectively.

Subset	Mean ± 1 SD	Median [5th, 95th]	п		
$1 \le \mu < 2$					
DS-AD	-0.00 ± 0.70	0.00 [-1.15, 1.09]	399		
DS	-0.12 ± 1.25	0.00 [-2.15, 1.63]	4070		
ZS	0.01 ± 1.78	0.00[-2.80, 2.98]	5682		
ALL	-0.04 ± 1.58	0.00 [-2.62, 2.62]	9752		
$2 \le \mu < 3$					
DS-AD	0.02 ± 0.82	0.00 [-1.10, 1.24]	463		
DS	-0.18 ± 1.27	-0.25 [-2.22, 1.80]	3376		
ZS	-0.10 ± 2.11	0.00 [-3.40, 3.40]	5102		
ALL	-0.13 ± 1.83	-0.17 [-3.06, 2.93]	8478		
$3 \le \mu < 4$					
DSCD	0.01 ± 1.39	0.00 [-2.26, 2.24]	442		
DS	-0.32 ± 1.73	-0.30[-3.03, 2.27]	826		
ZS	0.11 ± 2.53	-0.07 [-4.58, 4.04]	1490		
ALL	0.19 ± 2.28	-0.12 [-3.90, 3.54]	2316		

with nearly parallel DS measurements, not taking into account the specific cloud configuration and optical properties.

3 Results and discussion

The following subjects were considered to construct the Brewer-adjusted record of Belsk's Dobson measurements for the entire observation period (1963–2019): the focus was on the DS-AD type of the Dobson observations during the intercalibration campaigns, DS-AD observations were well calibrated throughout the observation period (within $\pm 1\%$ range), the inter-comparisons were carried out in the warm period of the year (usually in summer), and there was a seasonal difference between the Dobson and Brewer spectrophotometers, depending to some extent on the effective temperature and the different response to the stray light in these instruments. It is worth mentioning that BS64 will ultimately be the primary ozone monitoring instrument at Belsk.

The first step in the data homogenization was the correction of Dobson's ZS observations, taking into account the drift in relation to DS–AD values (Fig. 2). Application of the correction function according to Eq. (1) removed this shift (Fig. 2b). Moreover, Fig. 3a and b ensured that further correction to other types of the measurements was not necessary to eliminate long-term drift against the quality-controlled DS–AD subset. Thus, ZS-DS conversion charts proposed by



Figure 10. The ratio between the daily mean TCO_3 value based on the observations during the day with the slant TCO_3 smaller than 800 DU and daily mean TCO_3 comprising measurements with the slant ozone greater than 800 DU (points): the results are for DS measurements (a) and ZS measurements (b). The curve (in blue) represents the smoothed data.

Table 3. The monthly and yearly statistics (mean, standard deviation (SD), median, and the 5th–95th percentile range) of the differences between the Brewer-adjusted Dobson TCO₃ and original (Bass–Paur O₃AC at fixed temperature) TCO₃ for the period 1963–2019; *n* denotes the number of the TCO₃ daily means used in the calculations.

Month	п	$Mean \pm SD \\ (\%)$	Median [5th, 95th] (%)
January	1255	3.61 ± 0.81	3.68 [2.08, 4.78]
February	1287	1.51 ± 0.82	1.39 [0.35, 3.15]
March	1515	0.67 ± 0.34	0.63 [0.18, 1.26]
April	1497	0.27 ± 0.23	0.25 [-0.07, 0.69]
May	1577	-0.09 ± 0.19	-0.10 [-0.36, 0.21]
June	1460	-0.40 ± 0.16	-0.42 [-0.61, -0.15]
July	1594	-0.53 ± 0.15	-0.54 [-0.73, -0.33]
August	1548	-0.41 ± 0.18	-0.44 [$-0.67, -0.07$]
September	1520	-0.03 ± 0.23	-0.05 [-0.35, 0.35]
October	1546	0.51 ± 0.31	0.47 [0.10, 1.02]
November	1339	3.13 ± 1.16	3.40 [1.05, 4.71]
December	1251	4.48 ± 0.53	4.54 [3.44, 5.24]
Year	17 389	0.93 ± 1.68	0.25 [-0.59, 4.55]

Rindert (1973) were valid throughout the observation period, suggesting slight changes in the cloud characteristics at Belsk in the period 1963–2019.

The next step was to account for the temperature dependence of O₃AC based on the very recent O₃AC derived by Serdyuchenko et al. (2014). Application of Eq. (2) removed part of the bias between the Brewer and Dobson spectrophotometers for both DS and ZS subsets of the observations (Figs. 7–8). The BD monthly mean differences within the $\pm 1\%$ range occur in almost all months but not in months with low solar elevation at noon (November–December– January).

The last step of the homogenization was to eliminate the differences between the spectrophotometers found in periods of low solar elevation. These are probably related to the



Figure 11. Yearly means of the effective temperature at Belsk in December and July in the period 1963–2019.

presence of stray light in spectrophotometers causing an underestimation of TCO₃ values at low solar elevations. The correction for the differences in the stray light effect in the spectrophotometers was proposed using Eq. (3) to reduce the BD TCO₃ differences for low solar elevation. The stray light correction was not calculated separately for each instrument. However, Fig. 10 shows that the Brewer-adjusted TCO₃ values are only slightly sensitive to changes in slant TCO₃, i.e., within the min–max range between 0.99 and 1.01 derived from the smoothed profile of the ratio between TCO₃ values unaffected (slant TCO₃ < 800 DU) and affected by the stray light.

Table 3 provides the monthly mean values of the differences between the final and original TCO₃ values. The larger differences of about a few percentage points are found in the late autumn and early winter months. For the warm season (April–September) and the corresponding higher solar elevation at noon, the differences are only in the range of ± 0.5 %.

Figure 11 shows that in December there was a significant decline in the effective temperature by $\sim 4 \,^{\circ}$ C in the period



Figure 12. Time series of the averaged daily TCO₃ means for Belsk in the period 1963–2019 using the original data (Bass–Paur O₃AC at fixed temperature) and the Brewer-adjusted Dobson data: monthly means in December (**a**), monthly means in July (**b**), and yearly means (**c**). The red and blue curves show the smoothed pattern for the original data and the Brewer-adjusted Dobson data, respectively.

1963–2019 and a slight decline in July by $\sim 1 \,^{\circ}$ C. For the original TCO₃ data (archived in the PANGAEA database), such a decline caused the TCO₃ to be underestimated by $\sim 0.4 \,\%$ and $\sim 0.1 \,\%$ at the end of the time series compared to the TCO₃ values at the start, in December and June, respectively.

Figure 12a and b show the long-term variability in the monthly mean TCO₃ for December and July, respectively, derived from the original data and the data after application of all correction functions (i.e., the Brewer-adjusted Dobson TCO_3). The monthly means were calculated averaging the daily means from DS observations. If there were no such observations within the day, the daily means based only on ZS measurements were considered. The decline in the effective temperature (Fig. 11) does not change the variability of the long-term TCO₃ pattern. A constant upward shift is found in the colder months (e.g., Fig. 12a) and a slight downward shift in the warmer months (e.g., Fig. 12b). A constant shift (upward) is also seen in the yearly mean values calculated as the average of all monthly means for each year (Fig. 12c). Therefore, the trend estimates expressed in DU will be almost the same in the original and corrected datasets if the trends are calculated as differences from the reference TCO₃ value (e.g., the TCO₃ monthly means before 1980).

4 Data availability

The dataset used in this article is available on the PANGAEA repository at https://doi.org/10.1594/PANGAEA.919378 (Rajewska-Wiech et al., 2020).

5 Conclusions

The TCO₃ observations by the Dobson spectrophotometer at higher latitudes such as Belsk, with frequent non-ideal conditions for ozone observations, i.e., numerous cloudy days and low solar elevation at noon during the cold period of the year, should be subject to special quality checks prior to searching for the long-term trends and comparisons with satellite data. Frequent intercomparisons of Belsk's Dobson spectrophotometer, which were carried out in perfect weather conditions, showed the stability of the instrument throughout the observation period for the most reliable DS–AD observations. However, such observations were not possible for many days, especially in the cold season of the year. The data correction procedure is proposed to account for less accurate observations at low solar elevations, the presence of clouds, and the temperature sensitivity of ozone absorption.

The results of all intraday measurements for the period 1963–2019 were previously stored at the PANGAEA repos-

itory base with additional information including time of observation (UTC), cloudiness type, air mass, and description of the wavelength pair and observation type selected for each individual measurement (Rajewska-Więch et al., 2020). The present analysis shows that the Brewer-adjusted Dobson TCO₃ values are reliable for $\mu < 4$ or slant TCO₃ up to 1400 DU (Fig. 10).

Corrections according to Eqs. (1) and (3) are a linear function of air mass, so any user can easily calculate them based on archived air masses for all measurements. The effective temperature can be taken from the TEMIS database, and the correction for the temperature dependence of ozone absorption, given by Eq. (2), also requires simple calculations. Thus, it will be possible to use the homogenized Dobson data for comparative studies with other TCO₃ data sources.

The present analysis shows that the original data (archived in the PANGAEA repository) can be used in trend analyses based on the TCO₃ monthly and yearly means. The application of all proposed corrections to the original data ensures that the resulting smoothed time series exhibits a constant shift (upward for the cold season and much less downward for the warm season) relative to the original long-term pattern. This finding supports the validity of previous trend analyses of Belsk's Dobson data (e.g., Krzyścin and Rajewska, 2009; Krzyścin et al., 2013, 2014). The resulting Breweradjusted Dobson TCO₃ values are found to be in good agreement with the Brewer data. This will allow for the construction of the merged Dobson–Brewer time series as the Brewer observations will soon replace the Dobson measurements at Belsk.

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