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DLM estimates of long-term Ozone trends from Dobson and Brewer Umkehr profiles

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Abstract.

Six collocated spectrophotometers based in Arosa/Davos, Switzerland, have been measuring ozone profiles continuously since 1956 for the oldest Dobson and since 2005 for the most recent Brewer instrument. The datasets of these 2 ground-based triads (3 Dobsons and 3 Brewers) allow continuous intercomparisons and derivation of long-term trend estimates. In this study, the post-2000 ozone profile trends are estimated from the Dobson and the Brewer Umkehr time series, following a careful homogenization of the Dobson D051 dataset.

Mainly, two periods in the post-2000 Dobson D051 dataset show anomalies when compared to the Brewer triad time series: in 2011-2013, an offset has been attributed to technical interventions during the renewal of the spectrophotometer acquisition system; in 2018, an offset with respect to the Brewer triad and Aura MLS has been detected following a technical intervention on the spectrophotometer wedge. The Dobson D051 time series was homogenized using its difference to the Brewers triad N values time series and the ozone profiles were retrieved from the modified N values by optimal estimation method (OEM). Simultaneously, the Dobson D051 time series was optimized by NOAA relying on the NASA M2GMI model simulations to identify and correct instrumental artifacts in the record. Comparisons of the two homogenized data records show common correction periods and globally similar intensities of the N values corrections.

Long-term trends were estimated by Dynamic Linear Modeling (DLM) from Dobson ozone profile time series and, for the first time, from Brewer ozone profile time series. A positive trend of 0.2 to 0.5 %/year is estimated above 35 km, significant for Dobson D051 but lower and therefore non significantly different from zero at the 95% level of confidence for Brewer B040. As shown on the Dobson D051 data record, the trend seems to become significantly positive only in 2004. Moreover, a persistent negative trend is estimated in the middle and the lower stratosphere with different levels of significance depending on the dataset.

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1 Introduction

The stratospheric ozone layer is essential for its role in protecting the Earth's surface from harmful solar ultraviolet radiation. Stratospheric ozone depletion occurring during the second half of the twentieth century has been contained by the strict application of the Montreal Protocol and its amendments (MP1, 1987). While in the upper stratosphere (10–1 hPa, 32–48 km), ozone has started to show significant signs of recovery (e.g. Petropavlovskikh et al., 2019), in the lower stratosphere (147–32 hPa, 13–24 km), measurements show that ozone is still decreasing (Ball et al., 2018). Uncertainties remain for the middle stratospheric trends (32–10 hPa, 24–32 km) with different composites showing different changes giving a picture of a relatively flat trend with low significance (Ball et al., 2018).

Intensive discussions about the significance of the lower stratospheric trends and about the discrepencies between the magnitudes of the model simulated and the measured ozone trends are ongoing in the recent literature. Using TOMCAT 3-D CTM, Chipperfield et al. (2018) pointed to large interannual variability rather than an ongoing downward trend. Wargan et al. (2018) confirms the negative trend in the lower stratosphere (LS) in the northern hemisphere (NH) using DLM on MERRA-2 reanalysis. Sensitivity analyses by Ball et al. (2019) and Dietmüller et al. (2021) support the negative NH LS trends highlighting, for the former, the overestimated magnitude of the final years (-2018) anomalies by the models and, for the latter, the underestimated probability density function of the model trends, as causes for the bad accordance between the simulated and measured ozone LS trends. Orbe et al. (2020) associated the negative NH LS trends with a change in advection, describing a northward upwelling expansion associated with an enhancement of the downwelling over NH mid-latitudes. In this case, the discrepencies in magnitude between the LS trends retrieved from the measurements and from the models (M2GMI and MERRA2 reanalysis) are attributed to an imperfect simulation of the tropical convective processes and of the 2016 inversion of the QBO.

Multilinear regression (MLR) is widely and consistently used for ozone trend estimation. This is the dominant method in the recent and past trend estimate literature (e.g. Petropavlovskikh et al., 2019; Sofieva et al., 2021; Maillard Barras et al., 2020; McPeters et al., 1996a; Reinsel et al., 2002; Tummon et al., 2015; WMO, 1998; Staehelin et al., 2001, and references therein). Trend estimates are obtained by fitting a MLR function to the monthly mean ozone time series , presuming a linear dependence of the ozone content towards the explanatory variables and a linear increase or decrease of the ozone content over time. The sensitivity of the post-2000 trend magnitude towards the time range has been extensively discussed (e.g. Bernet et al., 2019; Dietmüller et al., 2021). Non monotonic post-2000 trends have also been reported in Arosio et al. (2019) where MLR trends are estimated from a merged SCIAMACHY, OMPS and SAGEII dataset on the 2003 to 2018 period. In their study, stratospheric tropical trends are shown to be negative during the 2004 to 2011 period and positive since 2012.

Trend estimates by DLM are recent in the literature. First reports are from Laine et al. (2014) who developed the DLM analysis for trend evaluation and applied it on a merge of SAGEII and GOMOS data records. They compare trend estimates by DLM to trend estimates by piecewise MLR, the latter being described in a companion paper by Kyrölä et al. (2013). They conclude that DLM is a robust method well suited for modeling ozone time series changes (see Section 4.2). Their results show a statistically significant turnaround in the ozone time series after 1997 at mid-latitudes in the 35 to 55 km altitude range and a more complex behavior of the ozone concentration than the description which can be made by a simple piecewise multilinear

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regression model. Consequently, stronger ozone variations (decrease or increase) are reported locally when estimated by DLM than by MLR. Ball et al. (2017) applied DLM on a Bayesian composite (BASIC) of satellites data records. The changes in ozone between 1998 and 2012 estimated using DLM indicate a clear and significant ozone recovery in the upper stratosphere (UpS). DLM has also been used to estimate trends in the LS based on the merged SWOOSH/GOZCARDS data records (Ball et al., 2018) as discussed previously. More recently, DLM trend estimates on SOS (SageII, Osiris and SAGEIII) merged satellite data record have been reported (Bognar et al., 2022) and indicate a clear UpS ozone recovery with varying turnaround years depending on the latitude, a decrease since 2012 in the NH upper/middle stratosphere, but without excluding a step in the Osiris dataset as a cause, and a persistent decrease in the tropical LS.

Dobson Umkehr ozone profile data record have been extensively used in the pre-1998 stratospheric trend estimates (Reinsel et al., 1989; Randel et al., 1999; Miller et al., 1995). Beginning in 1956 for the oldest, the Umkehr records were unique at that time since satellites records only became available in 1979 (McPeters et al., 1996b; Bhartia et al., 2013) and radiosondes, starting in 1960 (Smit et al., 2007), do not reach the UpS. Few studies based exclusively on Umkehr measurements report on post-2000 stratospheric ozone trends (Zanis et al., 2006; Park et al., 2013). Zanis et al. (2006) derived trends from the Arosa Dobson Umkehr dataset and reported statistically significant negative trends in the 1970 to 1995 period, and the first signs of a reversing trend in the lower and the upper stratosphere for the period 1996 to 2004. Since this turnaround was not statistically significant, the authors suggested that the dataset should be reevaluated at a future stage when more measurements become available. The Dobson Umkehr dataset was then corrected in 2008 for technical issues (Maillard et al., 2008) and ozone profiles were reprocessed from corrected observations (N values). The homogenized Umkehr time-series was used by Park et al. (2013) to derive trends using functional mixed models, and in the frame of the LOTUS project (Petropavlovskikh et al., 2019), which derived stratospheric ozone trends from improved and combined datasets (satellites, ground-based and models). The trends derived from the Umkehr datasets are in accordance with trends derived from other ground-based instruments for the pre-1997 period and the post-2000 period. Umkehr data corroborate also the satellite findings showing highly statistically significant evidence of declining ozone concentrations since the mid 1980s in the UpS and post-2000 positive trends ranging between 2.0% and 3.1% per decade in the UpS of NH mid-latitudes. The Umkehr data records are still extensively used for trend estimates along with datasets from other ground based techniques, satellites and models (Steinbrecht et al., 2017; Harris et al., 2015; Petropavlovskikh et al., 2019; Tarasick et al., 2019). However, to the authors' knowledge, no trend estimate including explanatory variables and based on the Brewer Umkehr data records has been reported in the literature so far. A study using simple linear regression applied to data from the Brewer 005 of Thessaloniki presented by Fragkos et al. (2018) reports 1997-2017 statistically significant positive trends above 35 km of 0.3%/year and non statistically significant trends below.

The dataset quality is of primary importance for trend studies, and multi-instrument comparison analyses is suited to assess the long-term stability of data records by estimating the drift and bias of instruments. A reprocessing and a homogenization of Dobson total ozone (TCO) data record is in progress with consideration of the effects of the relocation from Arosa to Davos (Stübi et al., 2021b) and the use of ozone absorption cross section from Serdyuchenko et al. (2014) (Gröbner et al., 2021). The complete homogenization of the Dobson (D051) Umkehr data record is described in this paper.

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The paper is organized as follows: the data sources used in this study are described in section 2, with a special focus on the Umkehr method description. In section 3, the complete homogenization of the Dobson D051 Umkehr data record is detailed and compared to the homogenization performed by NOAA in the frame of the ESA project WP-2190 (Garane et al., 2022). The trend estimate methods are described in section 4. DLM trend estimate results are presented and discussed in section 5, followed by conclusions in section 6.

2 Data Sources

2.1 Umkehr data records from Arosa/Davos

TCO and ozone profile measurements with Dobson (and Brewer) spectrophotometers were performed at Arosa from 1926 (and 1988) to 2021 and at Davos since 2012. For a detailed description of the Dobson and Brewer spectrophotometers, we refer to Stübi et al. (2021a, 2017a). The progressive relocalisation of the Dobson and Brewer triads from Arosa to Davos (13 km north of Arosa and 260m lower in altitude) between 2012 and 2021 is described and analyzed in Stübi et al. (2017b, 2021b). Umkehr measurements (see Section 2.2) are performed under clear sky and low cloud cover conditions twice a day since 1956 by Dobson spectrophotometers (Dobson D015 since 1956 and then Dobson D051 since 1988), 4–6 times per month by Dobson 101 since 1988 and by Dobson 062 since 1998. Dobson D051 performs fully automated Umkehr measurements since 1988. The Dobson Umkehr measurements are complemented by Brewer Umkehr measurements since 1988 with Brewer B040 and since 2005 with Brewers 072 and 156. Ozone profile Umkehr measurements initiated in 1956 at Arosa and continued since 2021 at Davos, Switzerland, form the longest continuous Umkehr measurement time series world-wide (Staehelin et al., 2018).

At Arosa, the Dobson D051 sat on a turntable in a conditioned hut maintained at 25-28°C. An aperture in the roof, which opened and closed according to solar zenith angle (SZA) and weather conditions, allowed zenith measurements. The continuous and automated measurements (2 min cycle) are interpolated to 12 nominal solar zenith angles (SZA) and profiles are retrieved using OEM from ground to 50 km. Manual Umkehr measurement started in 1968 with the Dobson D101 and in 1992 with the Dobson D062 as redundant measurements to check the stability of Dobson D051 . These are made two to three times each month since 1988 and 1998 in favourable weather conditions. The Dobson D062 and the Dobson 101 have been automated in 2012 and in 2013, respectively (Stübi et al., 2021b). They are located since 2021 in Davos in a common air conditioned container side by side with the Dobson D051, and measure Umkehr curves through a quartz dome. While the Dobson D051 was dedicated exclusively to Umkehr measurement until February 2013, the present set-up allows both direct sun and zenith Umkehr measurements with the three Dobsons. The Arosa/Davos Dobson instruments are regularly calibrated against the two European regional secondary reference Dobson instruments D064 from the Hohenpeissenberg Observatory (MOHp, Germany) and D074 from the Solar and Ozone Observatory in Hradec Králové (SOO-HK, Czech Republic) (Stübi et al., 2021b, Fig. 3).

The Brewer triad consists of two Brewer Mark II single-monochromator instruments, the Brewer B040 and the Brewer B072, and one Brewer Mark III double-monochromator instrument, the Brewer B156. The three instruments measure daily in Umkehr mode when the sun is at the 12 nominal SZAs. Since the operation of the first Brewer at Arosa in 1988, biennial calibrations have been carried out (Stübi et al., 2017a, Fig. 1) towards the traveling reference instrument Brewer 017 and,

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since 2008 towards the traveling reference instrument Brewer 185. The instruments of the Brewer triad underwent very few technical interventions and are in good agreement with the travelling references (TCO deviations <=1%, (Stübi et al., 2017a)). In particular, no technical issues are reported around 2011-2013 and 2018, which are data records periods considered in the frame of the Dobson D051 homogenization. However, sporadic instabilities in the Brewer B072 data record have been observed while no particular technical issues have been detected by the intercomparison procedures. The Dobson D051, the Brewer B072 and the Brewer B156 have been simultaneously relocalized from Arosa to Davos in September 2018 however with an effect on the TCO level within the instrumental noise (Stübi et al., 2017a, 2021b).

2.2 The Umkehr method

The Umkehr method is based on the measurement of the ratio of the zenith-sky UV intensities at two wavelengths in the UVB-UVA range from 300 nm to 330 nm (Huggins absorption band) subject to different strengths of ozone absorption. This ratio is changing as a function of SZA during sunset and sunrise. As the SZA is decreasing from 90° to 60°, the two intensities decrease, the shorter wavelength intensity decreasing more rapidly than the longer wavelength intensity. When the effective scattering height for the shorter wavelength is above the ozone layer, its intensity decreases less rapidly because of the absorption occuring mostly after the scattering event. The ratio shows therefore a maximum at high SZA called the Umkehr effect (Mateer, 1965).

2.2.1 Measurements by Dobson spectrophotometer

The logarithm of the ratio of the two wavelengths (R values) is converted to radiance using calibration tables and reported as N values (Fig.1a) in N-units for 12 nominal SZAs between 60° and 90°(60°, 65°, 70°, 74°, 77°, 80°, 83°, 85°, 86.5°, 88°, 89° and 90°). The nominal wavelength pairs used in a Dobson spectrophotomer are A: 305.5 & 325.4 nm, C: 311.45 & 332.4 nm, and D: 317.6 & 339.8 nm. Two narrow slits separate the respective wavelengths. The ozone profiles (Fig.1b) are retrieved from the measurements of the C pair intensity while the total column measurement requires a combination of 2 wavelength pairs (AD) (Stübi et al., 2021a).

The 12 N values (further called N curve) are screened for clear sky conditions and corrected for cloud influence using a nearby UV/VIS lux meter. This empirical correction is based on the relation between the UV/VIS intensity of clear days (within the same month, for each SZA) and the UV/VIS intensity variation during the cloudy N curve measurement (see Basher, 1982). This cloud correction is based on a uniform cloud layer and may fail for more complicated cloud structures. Haze correction is not included. For these reasons, only profiles retrieved from N curves without any cloud correction or with a small correction are considered for our study.

2.2.2 Measurements by Brewer spectrophotometer

The intensity of 8 wavelenghths (306.3, 310.1, 313.5, 316.8, 320.1, 323.2, 326.5, and 329.5 nm) are quasi-simultaneously measured for solar zenith angle changing from 60° and 90°. A holographic grating is used as dispersive element for the solar radiation passing then through narrow slits centered on the desired wavelengths. Mk II Brewer instruments use one single

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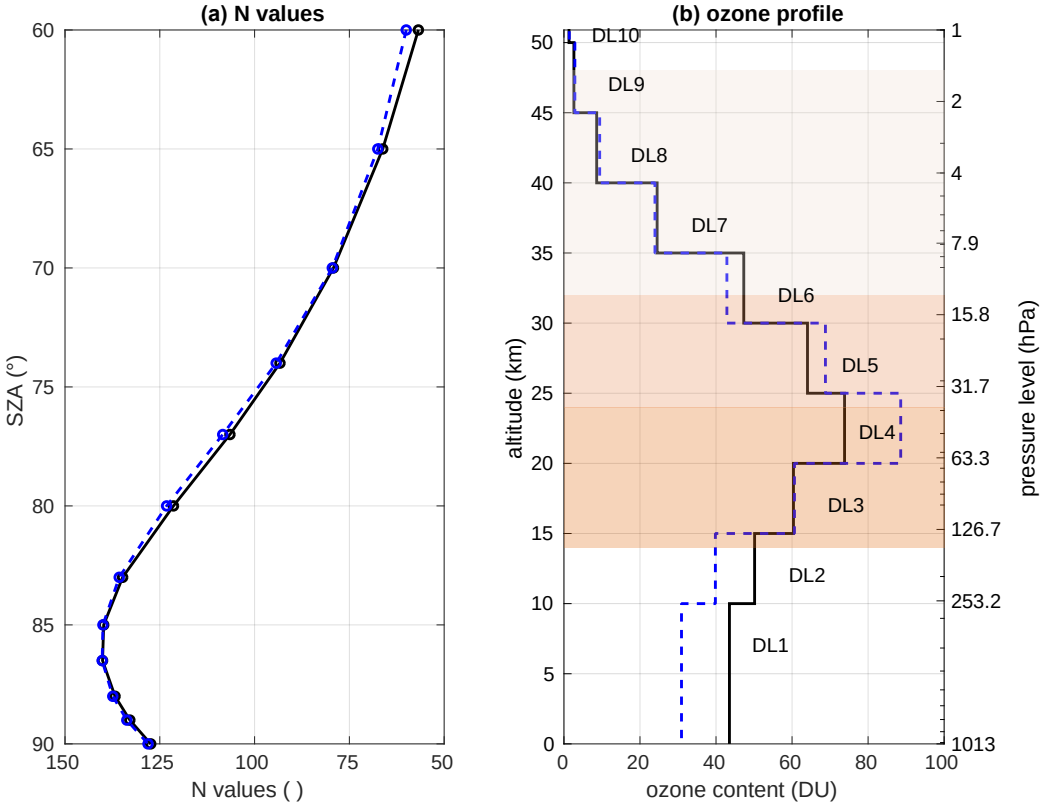

Figure 1. Two examples of N curves at 12 nominal SZAs and their corresponding retrieved ozone profiles in DU in function of altitude in km and pressure level in hPa. Altitude ranges of the 10 Dobson layers (DL) are indicated. Lower, middle and upper stratospheric ranges are displayed in shades of orange.

holographic grating and therefore only one dispersive element to separate the wavelenths. Mk III Brewer instruments are double monochromators that use two holographic gratings (Staehelin et al., 2003). The Umkehr ozone profile is commonly retrieved directly from the absolute wavelength intensities (photocounts at the selected wavelength) by OEM. For similarity with the Dobson Umkehr measurement, the intensity ratio of only two wavelengths are used : 310.05 nm of short set of wavelengths, and 326.5 nm of the long set of wavelengths. The data are flagged for clouds before the interpolation onto the 12 nominal SZA. The quality filter eliminates data points that fall outside a predefined error envelope determined by the range of natural variability and a mean offset.

Dobson and Brewer ozone profiles are retrieved by OEM (Rodgers, 2000). The Dobson Umkehr retrieval algorithm is described in Petropavlovskikh et al. (2005a) and the Brewer Umkehr retrieval algorithm has been adapted by Petropavlovskikh et al. (2005b) from the Dobson algorithm. The version of the code used in this study has been implemented by M. Stanek and

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can be found at http://www.o3soft.eu/o3bumkehr.html. Ozone profiles are given on ten layers between 0 and 50 km with a vertical resolution of 10-15 km. Dobson and Brewer Umkehr retrievals are using the same a priori profile, the ML climatology, described in McPeters and Labow (2012) formed by combining data from Aura MLS (2004-2010) with data from balloon radiosondes (1988-2010). The measurement error covariance matrices are diagonal with values between 0.16 to 0.8 N-units for Dobson and 0.6 to 2 N-units for Brewer. The Brewer observation errors have been estimated by the standard deviation of the 2005–2018 climatological difference of collocated and simultaneous N values measurements. The AVeraging Kernels (AVKs, not shown) show that for both instruments, the information below Dobson Layer (DL) 4, peaking at 20 km, is not independent. A generic stray light correction can be applied to reduce systematic biases in the Dobson Umkehr retrieved profiles (Petropavlovskikh et al., 2011).

The Dobson D051 Umkehr observations dataset is regularly archived at the World Ozone and Ultraviolet Radiation Data Centre (WOUDC, www.woudc.org). The Brewers are part of the eubrewnet (http://www.eubrewnet.org/eubrewnet), where raw data files are available for registered users.

2.3 NOAA Dobson Umkehr data records (Boulder and OHP)

The Umkehr data record from Boulder is automated using the NOAA WinDobson operational software (Evans et al., 2017) that schedules zenith sky observations at C-pair spectral channels during the morning and afternoon hours. The software uses the near-IR cloud detector to screen the Umkehr data for clear sky conditions. Screened observations are interpolated to 12 nominal SZAs and total column ozone information is added. Data processing includes checks of the retrieved ozone profiles for quality flags and against station climatological variability (+/-2 standard deviations). NOAA Dobson Umkehr operational ozone profile data are posted on the GML archive https://gml.noaa.gov/aftp/data/ozwv/Dobson/AC4/Umkehr/. The Umkehr observations are archived at WOUDC where the centralized data processing is done by the python-based version of the UMK04 processing software (https://github.com/woudc/woudc-umkehr). (Petropavlovskikh et al., 2022).

2.4 Aura MLS

The Microwave Limb Sounder (MLS) is a microwave limb-sounding radiometer on board the Aura Earth observing satellite, launched in July 2004. Ozone profiles are retrieved from Aura MLS radiance measurements at 240 GHz. Details about the instrument can be found in (Waters et al., 2006). In this study we use ozone profiles from the version 4.2 dataset given on 55 pressure levels from 1000 to 1e-5 hPa (Livesey et al., 2018). However, the useful vertical range for Aura MLS ozone leads us to only consider Aura MLS data from 10 to 75 km (in this range, the Aura MLS vertical resolution is about 2.5 to 4 km) for Aura MLS overpasses above Switzerland (46.82° N, 6.95° E, ±3° in latitude and ±5° in longitude). These ozone profiles are interpolated on the Umkehr pressure levels p_i and converted to DU following Ziemke et al. (2017):

$$X_{DU} = C * \bar{X} * (p_i - p_{i-1}) \quad (1)$$

with $\mathrm{C} = 0.00079 \mathrm{DUhPa}^{-1}\mathrm{ppbv}^{-1}$ and $\bar{\mathrm{X}}$ the ozone mean VMR in ppbv. Approximative height is given following the Umkehr layers definition (Petropavlovskikh et al., 2022).

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3 Homogenization of Dobson D051

While the 2008 homogenization dealt with the Dobson D015 to Dobson D051 transition (using one year of parallel measurements) and with correction of shaft encoder mispositioning on the raw data level, this study deals with a complete homogenization of the Umkehr Dobson D051 time series by comparison to the datasets of the 5 collocated instruments (2 Dobson and 3 Brewer spectrophotometers) on the N value level. The purpose is to detect common anomalies in the difference between Dobson D051 and each of the redundant measurements and to correct the Dobson D051 time series accordingly. However, a correction is only applied if it correlates with a technical issue in the metadata. If we cannot see any indication in the meta data for an instrumental drift, no correction is applied.

Figure 2 shows the time series of monthly mean ozone profile differences between Dobson D051 and the 5 collocated spectrophotometers. Only simultaneous measurements, flagged for bad weather conditions, volcanic eruptions, and for retrieval iterations higher than 3, are considered. The relative differences of the anomalies in % lie within +-15%. The comparisons with the Brewer instruments show a seasonal cycle with differences slightly bigger in summer than in winter (not shown; DL6: -2% in winter and +2% in summer). A similar behavior has been found by Gröbner et al. (2021) when comparing TCO from Dobsons to Brewers. Note that the annual cycle is not visible on the representation of deseasonalised anomalies as in Figure 2.

If we focus on the post-2000 period, where we have a precise chronology of the technical interventions, several systematic anomalies can be attributed to technical issues of the Dobson D051 (periods in black frames in Figure 2):

- before 2003 for the altitude range below 30km: the Dobson D051 ozone values are higher than the values measured by the collocated instruments below 20km and lower between 20 and 30 km
- in winter 2010 above 40 km, the Dobson D051 ozone values are higher than the values measured by the collocated instruments
- between 2011 and 2013 in most part of the altitude range, the Dobson D051 ozone values are lower than the values measured by the collocated instruments
- after 2018, the Dobson D051 ozone values are higher than the values measured by the 3 collocated Brewer instruments.

The comparison of Dobson D051 with the collocated Dobsons around 2014 and after 2018 are to be taken with caution due to the very limited number of measurements of Dobson D051 in 2014 and of Dobson D062 and Dobson D101 during these periods.

Table 1 summarizes the Dobson D051 problematic periods, the technical issue reported at these periods, and the time ranges and redundant datasets used for the offsets determination.

When systematic for each pair of instruments and if related to an instrumental issue, the detected Dobson D051 problematic periods are shifted according to the mean difference with the 3 Brewers or the 2 Dobsons datasets before and after the problematic periods (periods of 2 years are considered). The homogenization is performed on the raw data level (N values) and the ozone profiles are then retrieved from the corrected N values.

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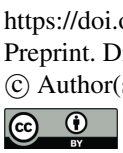

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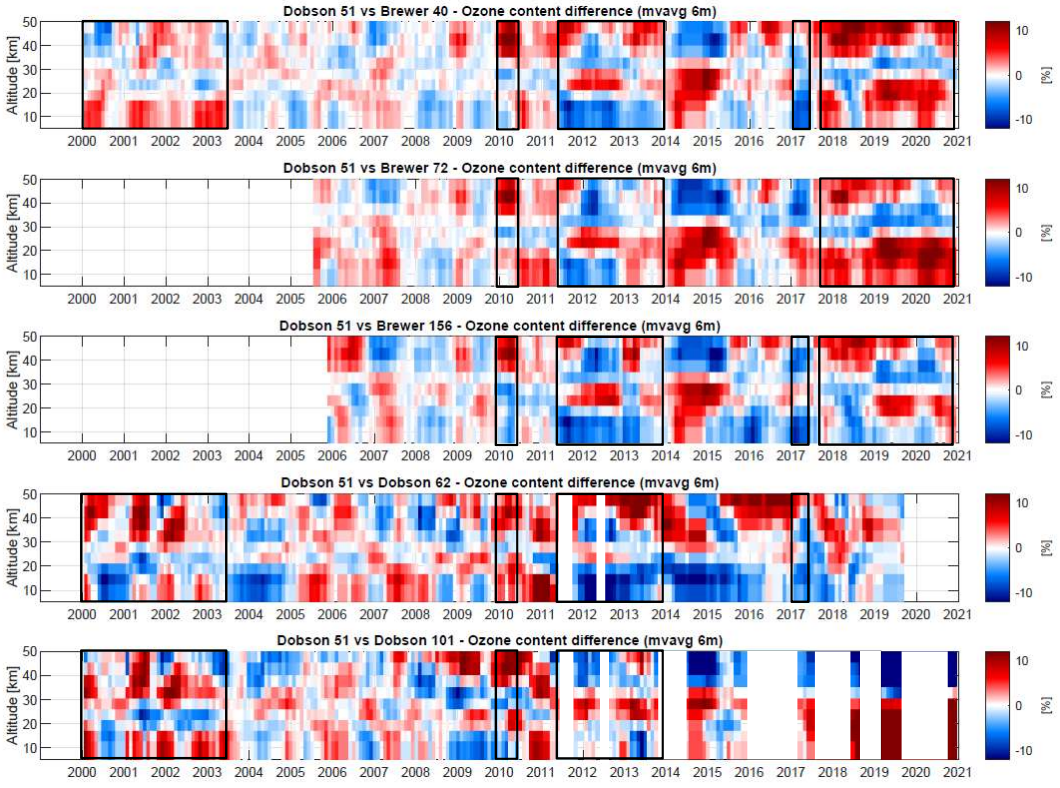

Figure 2. Monthly mean time series of the ozone profiles relative differences for each of the 5 spectrophotometers with respect to D051. The time serie are deseasonalized and smoothed by a 6 months moving average.

For each period that requires a correction (black frames in Fig. 2) we apply to the N values a SZA dependent offset which is constant over the period to be corrected. The offset is calculated such that the difference averaged over the period and over the reference instruments (2 Dobsons in 2003 or 3 Brewers after 2011) matches the difference averaged over 2 years before and 2 years after the period and over all reference instruments (see Fig. 3):

$$\Delta_{SZA} = mean(\Delta 1_{SZA}, \Delta 3_{SZA}) - \Delta 2_{SZA} \tag{2}$$

$$N2^{corr}_{SZA} = N2_{SZA} - \Delta_{SZA} \tag{3}$$

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Dobson D051 problematic period	Technical issue	Time range of homogenization	Time range used for the offset determination	Redundant datasets used for the offset determination	Comment
1988	D15 to D51	Before 1988.01.01	1987.01.01 - 1988.01.01 and 1988.01.01 - 1989.01.01	D015 and D051 simultaneous measurements	
2003	Intercomparison and new RtoN table New RtoN table considered	Before 2003.07.19	2001.07.19 - 2003.07.19 and 2003.07.19 - 2005.07.19	D062 and D101 mean values	
2010	-	2010.01.01 - 2010.06.30	-	-	Does not correspond to any technical issue. Period limited to 6 month. Not corrected.
2011-2013	New electronics (2011.03.21) New Qlever motors (2012.02.15) New software 3V3 (2013.03.26)	2011.04.01 - 2013.04.01	2009.04.01 - 2011.04.01 and 2015.04.01 - 2017.04.01	B040, B072 and B256 mean values	2014 not considered (number of measurement low and problematic period). Refurbishment of the electronics (HV, motors, feedback loop, amplification board) and position of Q2-lever in function of the room Temperature.
2018	New wedge steel band (2018.05.06) IC(2018.08.07-17): adjustments on optics AROSA to DAVOS (2018.09.28)	Before 2018.05.01	2016.05.01 - 2018.05.01 and 2018.05.01 - 2020.05.01	B040, B072 and B256 mean values	The optical attenuator consists of a moving neutral-density filter (the optical "wedge") attached to a graduated rotating disc (R dial). The wavelength pair selection is achieved by rotating a pair of quartz plates (Q1 lever, Q2 lever) through which the light beam passes.

Table 1. Dobson D051 homogenization description: problematic periods, technical issues, time ranges for homogenization and for offset calculation, used redundant datasets, details of technical issues

Δ_{SZA} is the offset between the 3 Brewers mean N values and the Dobson D051 N values for each SZA, $\Delta 1_{\mathrm{SZA}}$ and $\Delta 3_{\mathrm{SZA}}$ are the difference between the 3 Brewers mean N values and the Dobson D051 N values before (period P_1) and after (period P_3) the Dobson D051 problematic period (period P_2). All values are averaged over 2 years periods. $\mathrm{N2}_{\mathrm{SZA}}^{\mathrm{corr}}$ is the corrected N value in period P_2.

In case of a step in the time series (e.g. in July 2003 and in May 2018), the period P_2 does not exist and should not be considered in Fig. 3. The corrected N value $\mathrm{N2}_{\mathrm{SZA}}^{\mathrm{corr}}$ of period P_1 is then obtained following equations 4 and 5.

$$\Delta_{SZA} = \Delta 1_{SZA} - \Delta 3_{SZA} \tag{4}$$

$$N1_{SZA}^{corr} = N1_{SZA} - \Delta_{SZA} \tag{5}$$

In parallel, a homogenization and a correction for the stray light effect of the same Dobson dataset has been performed by NOAA, using comparison with the M2GMI model on the N values level. A summary of the homogenization method is presented here, for details on the method and for the description of the stray light correction, we refer to Petropavlovskikh et al. (2022).

The NASA Global Modeling Initiative chemistry transport model (GMI CTM) is an off-line model driven by MERRA2 meteorological reanalysis (Gelaro et al., 2017). M2GMI output is driven by a specified dynamics simulation instead of using MERRA-2 meteorology (Orbe et al., 2017). The ozone and temperature profiles are simulated by the M2GMI model for the Arosa station location. The simulated temperature profile is used for accounting the temperature dependence of the ozone

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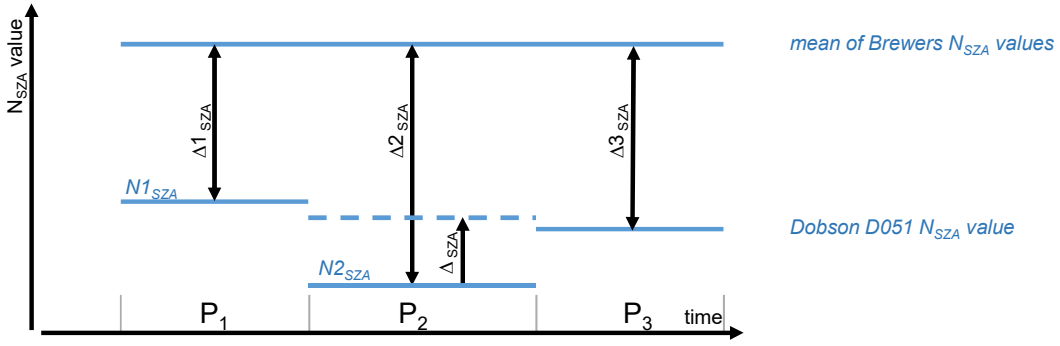

Figure 3. Schematic of the Dobson D051 MeteoSwiss (MCH) homogenization principle

cross section and allows the model to better fit to the day-to-day variability of the N values. The Umkehr retrieval forward model is forced to match this reference and outputs Umkehr N values simulated for an idealized Dobson instrument. For each SZA, differences between simulated and measured Umkehr N-values are averaged over the time between two consecutive calibrations (performed at each intercomparison) of the Dobson D051 to create an empirical correction for the Umkehr curve simulated by the forward model. This correction is then applied to the N values curves which are retrieved by OEM to get optimized ozone profiles. An iterative modification of the N value correction is performed in order to minimize the mean bias to Aura MLS in the 2005–2018 period. While the first iteration of the homogenization remove artificial steps in the Umkehr ozone profile records, the iterative part reduces the bias relative to other ozone observing systems.

The NOAA homogenization has been developed to remove artificial steps in the NOAA Umkehr ozone profile records and to reduce the bias relative to other ozone observing systems. Our approach is different in that the homogenization process aims to remove artificial steps in the Dobson D051 Umkehr profiles record by ensuring the independence (in absolute values) of the datasets towards the collocated instruments datasets.

Figure 4 shows the time series of the N values correction as a function of SZA as determined by the NOAA homogenization (Fig. 4a) and by the MCH homogenization (Fig. 4b, this study). For comparison purpose, the NOAA correction values have been offsetted with their mean difference after 2018.

The main differences between the two homogenizations are the variability of the corrections values and the correction of the volcanic eruptions periods. The high variability of the NOAA N values correction comes from the contribution of the stray light effect correction to the total N values correction. This is not corrected for in our N value homogenization. The years around 1982 and 1992 are periods of volcanic eruptions (El Chichon and Pinatubo) which are corrected by the NOAA homogenization but flagged in the MCH homogenization as the Umkehr retrieval does not account for the change in atmospheric scattering due to aerosols injection (Petropavlovskikh et al., 2022). For the 1988 to 2003 period, both homogenizations differs for the 77-83°

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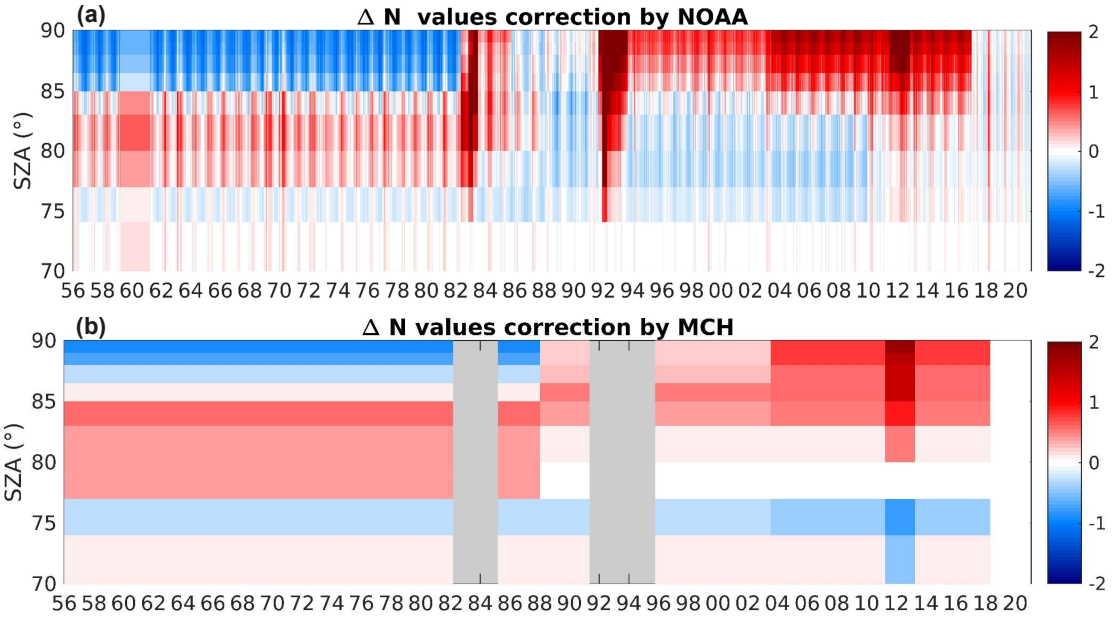

Figure 4. Monthly mean time series of the N values correction (a) for the NOAA and (b) for the MCH homogenization of Dobson D051 dataset. Volcanic eruptions periods (grey shaded area) are not corrected by the MCH homogenization.

SZAs. Otherwise, the correction amplitudes are similar, and their occurrences coincide within a few months in January 1988, in July 2003, in April 2011 to 2013, and in 2010; this is remarkable given the differences in the detection method. Note that the 2010 six-months step has been chosen to be left uncorrected in the MCH homogenization due to the absence of confirmed technical issue at that time. In 2017/2018, the start date considered for the NOAA homogenization is January 2017, while the start date considered by the MCH homogenization is May 2018 with the probable effects of the wedge steel band replacement on the measurements.

However, while both corrections of the N values looks similar, small differences in the N curve shapes can lead to larger differences in the ozone profiles due to the non linear relationship between the N values and the ozone values (see Fig.1 for an example).

In order to evaluate the effects of both homogenization on the Dobson D051 time series, monthly mean relative difference to Aura MLS data record are plotted in Figure 5 for 2 altitude levels i.e. DL5 (25 km) in MS and DL8 (40 km) in UpS. The relative difference of the Brewer B040 time series is also shown for the same layers.

The Brewer B040 relative difference shows a constant offset to Aura MLS but clear anomalies in 2012 and 2013 in DL5. The Dobson D051 optimized by NOAA shows a very good accordance with Aura MLS both in DL5 and DL8. The small overall offset value is a result of the optimization procedure which includes an iterative accordance of the 2005–2018 mean towards Aura MLS. No clear offset in the difference to Aura MLS between the NOAA and the MCH homogenized record

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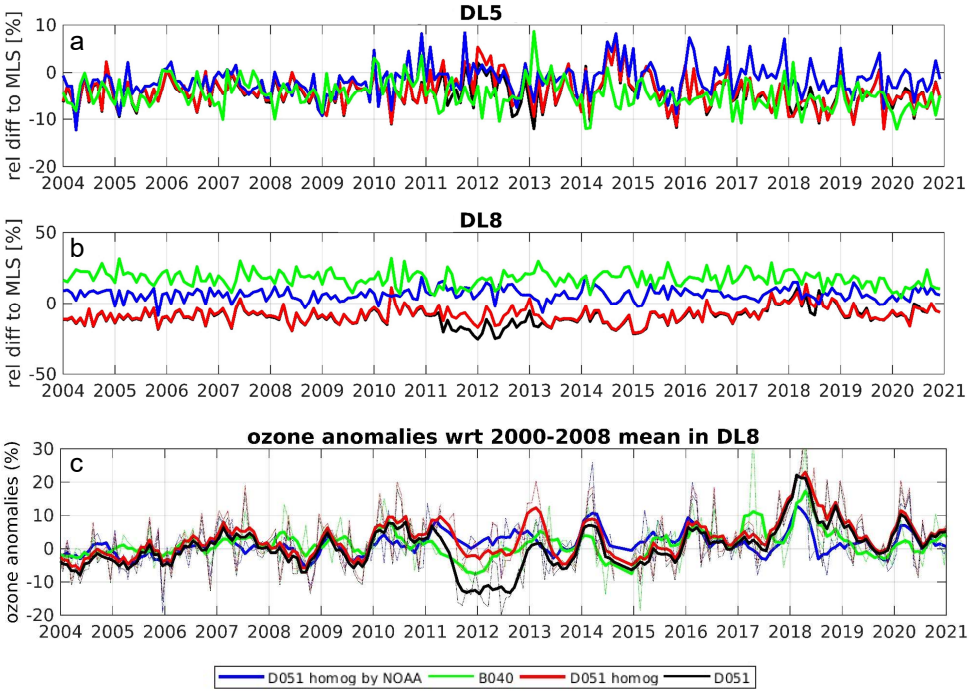

Figure 5. Monthly mean ozone content relative difference to Aura MLS for Dobson D051 as measured (black), Dobson D051 NOAA optimized (blue), Dobson D051 MCH homogenized (this study, red) and Brewer B040 (green) deseasonalized time series in (a) DL5 and (b) DL8. (c) Time series of ozone anomalies towards their 2000-2008 mean for the same ground-based datasets in DL8.

is reported in DL5. However, the slight underestimation of the MCH homogenization since 2017 seems to match the Brewer B040 difference to Aura MLS in DL5. After 2017, the relative difference to Aura MLS of D051 homogenized by MCH and of the collocated B040 is within -5% to -10% while the D051 homogenized by NOAA lies within -2% of Aura MLS. A clear correction of the 2011-2013 period is visible in DL8. Except for the respective MCH and NOAA homogenized datasets mean offsets to Aura MLS, a slight overestimation of the NOAA homogenization is visible in 2012 and 2013. However, the Brewer B040 relative difference to Aura MLS is also slightly smaller during this time range, when the Brewer instrument had not undergone any technical interventions. This is particularly visible on the anomalies time series of B040 in Fig. 5(c). As the MCH homogenization relies on the Brewer collocated datasets, it allows to take into account the local variability of the ozone DL8 content that the M2GMI model, base for the NOAA homogenization, probably does not consider.

In addition to the step correction in May 2018, the MCH homogenized dataset seems to show an anomaly in year 2018, as the collocated Brewer B040 dataset. The NOAA homogenization considering a correction in 2017, the 2018 anomaly is taken into account in the post-step period. The MCH homogenization considering a correction in May 2018, the 2018 anomaly is

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taken into account in the pre- and post-step period. As a result, the calculated offsets differ, with a probable overestimation of the offset for the NOAA homogenization and a very small offset correction for the MCH homogenization.

4 Trend estimate methods

4.1 MLR trend estimate

Trends are estimated by fitting a multi-linear regression (MLR) function to the monthly mean ozone time series considering two piecewise linear ramps (PWLT) starting in 1970 and in 2000. Trend profiles are obtained by considering one independent monthly mean time series for each pressure level. The results are given as a difference in DU to the 1970–1980 and of the 2000–2010 means. The explanatory variables represent sources of geophysical variability with known influence on stratospheric ozone, including the quasi-biennial oscillation[1] (QBO) at 30 and 10 hPa, the 10.7 cm solar radio flux describing the 11-year solar cycle[2] (SOL), the El Niño–Southern Oscillation[3](ENSO), the North Atlantic Oscillation[4] (NAO), the Stratospheric Aerosol Optical Depth[5] (SAOD) and Fourier components representing the seasonal cycle (annual and semi-annual variations). All data points are considered with equal weights, and the uncertainty of the fit parameters is estimated from the regression residuals. Residual autocorrelations are accounted for by applying a Cochrane-Orcutt transformation to the model (Cochrane and Orcutt, 1949).

4.2 DLM trend estimate

Dynamic linear Modeling (DLM) allows the determination of a non-linear time-varying trend from a monthly means time series. This is a Bayesian approach regression which fits the data time series for a non-linear time-varying trend, regression coefficients from explanatory variables and seasonal and annual modes, considering their uncertainties and an autoregressive component. The trend is allowed to smoothly vary in time and its degree of non-linearity is inferred from the data, as well as the turnaround period. We use the code by Alsing (2019) which is a Python implementation of the formalism introduced by Laine et al. (2014) and we refer to these publications for a detailed description of the DLM principles. The used model considers standard regression components, allows a variability of the sinusoidal seasonal modes and includes the autoregressive (AR1) correlation process with variance and correlation coefficient as free parameters in the regression. The same five explanatory variables as in the MLR are used in the trend estimate: QBO at 30hPA and 10hPa, SOL, ENSO and SAOD NH values. The estimation of the posterior uncertainty distribution is performed with the Markov chain Monte Carlo (MCMC) method, and considers the uncertainties on the regression components, on the seasonal cycle, on the autoregressive correlation and on the non-linearity of the trend.

[1] from *http://www.spaceweather.gc.ca/solarflux/sx-5-mavg-eng.php*
[2] from *http://www.spaceweather.gc.ca/ solarflux/sx-5-mavg-eng.php*
[3] from *http://www.esrl.noaa.gov/psd/enso/mei/*
[4] from *https://climatedataguide.ucar.edu/ climate-data/hurrell-north-atlanticoscillation- nao-index-station-based*
[5] from *https://asdc.larc.nasa.gov/project/GloSSAC/GloSSAC_1.0*

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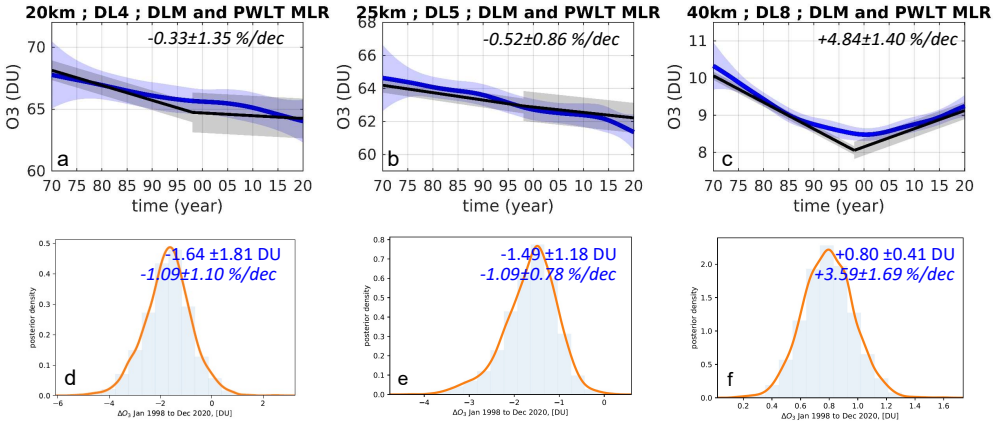

Figure 6. (a-c) DLM and MLR trend estimates in %/decade $\pm$ 2σ of Dobson D051 dataset for 3 DL between 20 and 40 km, the shaded areas show the 2σ uncertainties, (d-f) the distribution of the DLM trend estimates is given by the kernel density estimation (KDE) for the same 3 DL in the 1998–2020 time range in DU $\pm$ FWHM.

Figure 6 shows the long-term trend estimates from the Dobson D051 dataset by DLM (in blue with $\pm$2 sigma uncertainty shaded area) and by MLR (PWLT, in black with $\pm$2sigma uncertainty shaded area) for the same explanatory variables at 3 altitude levels.

Overall trends are similar but differ over short timescales because of their representation of the nonlinearity of the changes in the data record. The advantage of DLM lies in the estimation of a smoothly varying trend without assuming any shape. The inflection year depends on the method: while the inflection point is fixed by the MLR PWLT (1998 in this case, see Petropavlovskikh et al., 2019), the inflection year is retrieved by the DLM and results in year 2002 for the Dobson D051 dataset above 28 km. The maximum of the $\triangle$O3 1998-2020 KDE (kernel density estimation) should be compared to the linear trend value over the same time period (22 years), while the 95% level of significance, represented by the fraction of the KDE above/below zero, slightly differs from the MLR uncertainty estimates. In the LS, for DL4 (Fig 6a and d), the post-2000 MLR trends values are -0.33$\pm$1.35%/decade. The DLM KDE shows a maximum at -1.64 DU and a FWHM (=2.4 sigma for normal distribution) of 1.81 DU, which means a mean trend of -1.09$\pm$1.10%/decade. MLR estimate is non significantly different from zero at the 95% confidence level while DLM estimate is negative barely significant at the 95% level. In the MS, for DL5 (Fig 6b and e), the post-2000 MLR trends values are -0.52$\pm$0.86%/decade. The DLM KDE shows a maximum at -1.49 DU and a FWHM of 1.18 DU, which means a mean trend of -1.09$\pm$0.78%/decade. MLR estimate is non significantly different from zero at the 95% confidence level while DLM estimate is significantly negative at the 95% level. In the UpS, for DL8 (Fig 6c and f), the post-2000 MLR trends values are +4.84$\pm$1.40 %/decade and the DLM KDE shows a maximum at +0.80 DU and a FWHM of 0.41 DU, which means a mean trend of +3.59$\pm$1.69%/decade. Both are significantly positive at the 95% confidence level.

In case of high annual variability, a DLM trend estimate in %/decade may be significant while a MLR trend estimate may be non significant for the same considered period. The given DLM trend value in %/decade is an average of the % change

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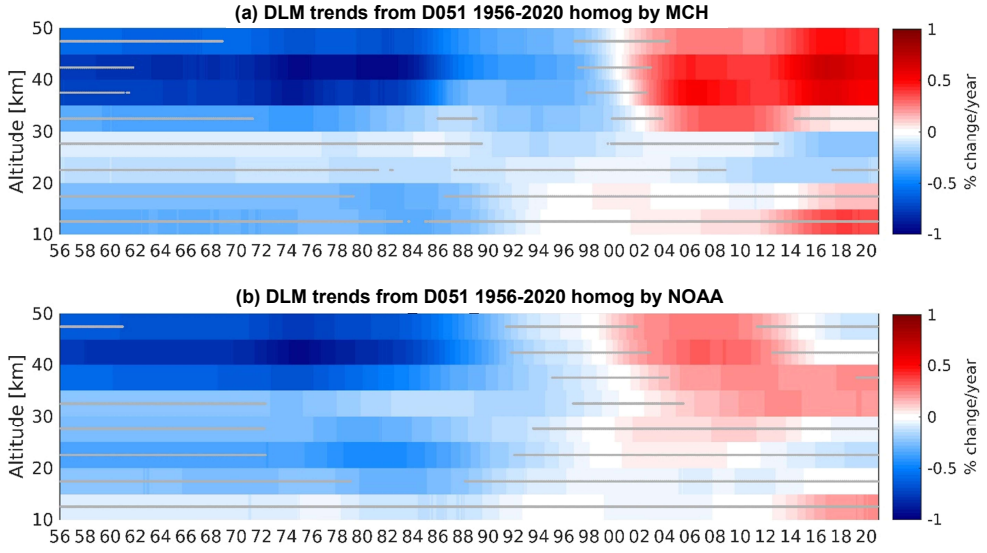

Figure 7. DLM trend estimates in %/year of Dobson D051 1956-2020 from (a) MCH homogenised and (b) NOAA optimized data records. Grey lines indicate trend estimates non significantly different from zero at the 95% confidence level.

per year. In a DLM estimate, only a limited number of values of % change per year may be influenced by an inhomogeneity, whereas the same inhomogeneity, especially when occurring at the boundaries of the time period considered, may influence the MLR trends significantly.

5 Results

Figure 7a and b shows the DLM trend estimates derived from the Dobson D051 record as homogenized by MCH (a, this study) and by NOAA (b). The trends values are given in % change per year for each altitude level between 10 and 50 km. Positive and negative trends are shown with varying intensities of red and blue respectively. The grey lines indicate trend estimates non significantly different from zero at the 95% confidence level.

The upper stratospheric (UpS, DL7-10, 10–1 hPa, 35–50 km) trend estimates are significantly negative between 1965 and 1997 on Figure 7a and before 1997 on Figure 7b. The mean negative trend estimates are -5 %/decade (mean value of the 1965-1997 UpS trends). Both records show then a transition period until 2003 with non-significant UpS trend estimates. The post-2003 UpS trends are significant and positive, up to 2020 for the MCH homogenized Dobson D051 record and until 2013 for the NOAA homogenized Dobson D051 record. The mean positive UpS trends are 3.6 %/decade on Figure 7a (mean value of the 2003-2013 UpS trends) and 2.1 %/decade on Figure 7b (mean value of the 2003-2013 UpS trends). Note that due to the large AVK of the Umkehr measurement, the ozone and trend informations in DL8 and DL9 are not independent. In the middle stratosphere (MS, DL5&6, 24–32 km), both homogenized records show a negative trend, persistent and significantly

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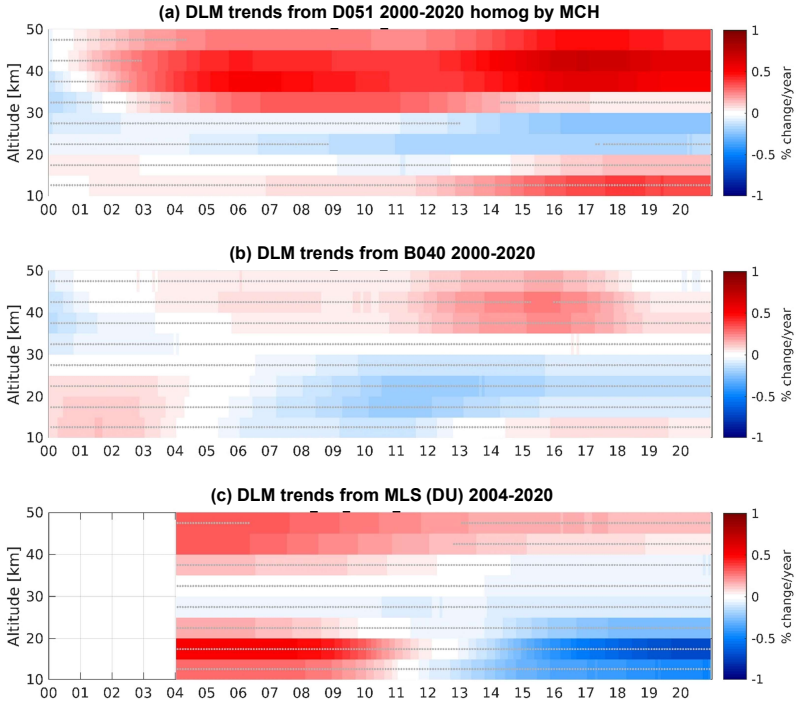

Figure 8. post 2000 DLM trend estimates in %/year from (a) Dobson D051, (b) Brewer B040 and (c) Aura MLS data records.

different from zero at the 95% confidence level since 2012 for the MCH homogenized Dobson D051 data record but slightly positive between 2002 and 2010 and non significantly different from zero at the 95% confidence level for the NOAA optimized data record. The lower stratospheric (LS, DL3&4, 14-24 km) trend estimates are non significantly negative before 1996 but significantly negative between 2008 and 2018 for the MCH homogenized data record and non significantly negative for the NOAA homogenized Dobson D051 record.

Again due to the AVKs width of the Umkehr profiles, the ozone content information of DL2 partly overpasses the LS as usually defined (see representation of shaded areas in Figure 1b). The same consideration is true for the DL6 and the MS. The lower part of the LS and the upper part of the MS trends may be aliased by upper tropospheric respectively upper stratospheric information.

Post-2000 trend have been estimated on the 3 Dobson and the 3 Brewer MCH Umkehr data records. The trends estimates of one of the Dobsons (D051) and one of the Brewers (B040) are represented in Figure 8 a and b in % change per year for each altitude level between 10 and 50 km.

The post-2000 trends show similar features for the two Swiss spectrophotometers:

- a positive trend of 0.2 to 0.5 %/year above 35 km, significant for Dobson D051 (and for Dobson D062 and Brewer B156 not shown) but lower and therefore non significantly different from zero at the 95% level of confidence for Brewer B040 and

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Dobson D101. Despite differences in the trend estimates intensities, an overall picture of a UpS positive trend after 2002 is shown.

- a persistent negative trend in the middle and the lower stratosphere with different levels of significance depending on the dataset but mostly non significantly different from zero at the 95% confidence level except for Dobson D051.

Significant UpS positive trends are estimated on the Aura MLS satellite data record (Figure 8c) however non significant since 2013. Signs of negative trends in the lower altitudes are also observed although not significant: DLM trend estimates are persistently negative in the MS and negative in the LS since 2012.

Both the Boulder D067 and the OHP D085 have been homogenized by NOAA towards M2GMI (Petropavlovskikh et al., 2022) and both data records are used for MLR trend derivation in LOTUS (Godin-Beekmann et al., 2022). D067, D085 (not shown) and Dobson D051 homogenized by NOAA show similar upper stratospheric post-2000 trends i.e. a statistically significant positive trend followed by a non significant negative trend, with a transition in 2014 for D067, in 2016 for Dobson D051 and in 2017 for D085. The LS trends are non significant and are not persistently negative.

6 Conclusions

Data records of six collocated spectrophotometers were inter-compared on the observation data level and on the ozone profile level in order to detect anomalies. The MCH Dobson D051 Umkehr data record has been homogenized on the observation data level by comparison to the collocated Brewer triad data record and to the redundant Dobson data records. In parallel, a second homogenization of the same Dobson dataset was performed by NOAA, using comparison with the M2GMI model on the raw data level as well. Both homogenizations result in similar magnitudes of N values corrections relative to the post 2018 values if we exclude the volcanic eruption periods for which corrections are performed by the NOAA optimization process but during which data are not considered in the MCH homogenization. On the other hand, by relying on the collocated Brewers datasets, the MCH homogenization accounts for the local variability of the ozone layers content in the 2011-2013 period and results in a weaker correction of the data record for this period.

Even if only slightly different, the homogenizations of the raw data can produce significant differences in ozone profiles and, therefore, in the long term trend estimates. When compared to Aura MLS and to Brewer B040, both homogenizations differ in the ozone profiles level in the UpS, especially for the period 2017-2019.

Trends of the ozone profile time series have been estimated by DLM from the Dobson and the Brewer spectrophotometers datasets. The post-2000 trends show similar features namely a positive trend of 0.2 to 0.5 %/year above 35 km in the UpS, significant for Dobson D051 but lower and therefore non significantly different from zero at the 95% level of confidence for Brewer B040, and a persistent negative trend in the MS with different levels of significance depending on the dataset. The DLM trend estimates from Dobson D051 show a significant persistent negative trend in the MS and supports also the mention of a persistent negative trend in NH LS when measured by ground-based instrument, considering, however, that the trends estimates in the upper part of the MS and in the lower part of the LS are aliased by the large AVKs of the Umkehr profiles.

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DLM trend estimates derived from Aura MLS show similar features in the UpS and the MS as estimates from the ground-based Dobson and Brewer spectrophotometers. However, a transition from non significant positive to non significant negative trends in the LS remains unexplained.

While significant positive trends have been estimated in the UpS since 2004 from the MCH homogenized Dobson D051 dataset, the trend estimates from the NOAA optimized data record appear to show a transition from significant positive to non-significant negative/zero values above 40 km in 2016. Further investigation will be needed to confirm this transition and exclude 2017 as a problematic period in the NOAA homogenization.

Both homogenization approaches considered in this study are relevant and significantly improve the Dobson D051 data record. However, inconsistencies in the level of significance of the Dobson D051 trend estimates are noticed and should be attributed to the remaining differences left by the homogenizations in the data records.

Data availability. The as measured Dobson D051 dataset is available at WOUDC. The NOAA homog Dobson D051 dataset is available at https://gml.noaa.gov/aftp/data/ozwv/Dobson/AC4/Umkehr/Optimized/Daily/ARO/. The MCH homogenized Dobson D051 dataset, the Dobson D062, Dobson D101 and the Brewers Umkehr datasets can be obtained from EMB on request. The MLS ozone dataset is available from the NASA Goddard Space Flight Center Earth Sciences Data and Information Services Center (GES DISC) at http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/MLS/index.shtml.

Author contributions. E.M.B. is responsible for the Umkehr ozone measurements with the Arosa/Davos Dobson and Brewer spectrophotometers, performed the data analysis and prepared the manuscript. A.H. and R.S. contributed to the interpretation of the results. A.J. performed the 2011-2013 homogenisation and the first DLM trend derivation. H.S. is responsible for the Arosa/Davos Dobson and Brewer spectrophotometers. I.P. and K.M. performed the NOAA homogenization of D051. M.S. implemented the Umkehr Brewer retrieval algorithm. L.F. is responsible for the Aura MLS measurements. All co-authors contributed to the preparation of the manuscript.

Competing interests. The authors have no competing interests.

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