

## Response to reviewer 1 comments and suggestions.

### General comments:

The authors present one of the longest series of measurements of the total column of ozone globally. They also discuss the conditions and the procedures which ensure the high quality of the measurements. The scientific value of the presented dataset is high, and the manuscript is within the scope of the journal.

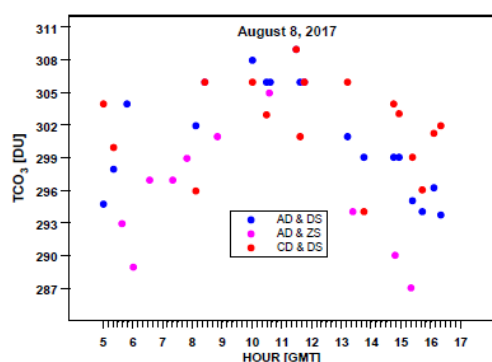
What I mainly miss, is a section wherein the authors would quantify the uncertainties of the final dataset. Uncertainty budget is of exceptional importance for anyone who would use the data. Thus, I strongly recommend that the authors should quantify the overall uncertainty and add the corresponding section.

The new section is added “2.4 Uncertainty of the Brewer adjusted Dobson  $\text{TCO}_3$ ” describing uncertainties in the data. (line. 253-281 in the revised manuscript).

### 2.4 Uncertainty of the Brewer adjusted Dobson $\text{TCO}_3$

Typically, daily  $\text{TCO}_3$  averages were archived based on a few measurements around local with nominally best quality. 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 the observation is not always possible because of weather conditions (clouds) and during low solar elevation. At the beginning of the  $\text{TCO}_3$  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  $\text{TCO}_3$  observation. On such days, there were many, almost simultaneous observations with different instrument settings. For example, Figure 9 shows the  $\text{TCO}_3$  measurements at Belsk on August 8, 2017, planned to calculate differences between successive  $\text{TCO}_3$  values. During the day, 46 observations were made for the following instrument settings: AD&DS, AD&ZS, and CD&DS.

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295 Figure 9. The Brewer adjusted TCO<sub>3</sub> values measured at Belsk on August 8, 2017 for different settings of the Dobson spectrophotometer.

The difference between two consecutive TCO<sub>3</sub> values sometimes exceeded 10 DU, even if the measurements were taken within 10 minutes. It is impossible for TCO<sub>3</sub> to change rapidly over such a short time scale taking into account natural variability of ozone in the stratosphere. The statistics of TCO<sub>3</sub> differences between the almost simultaneous Dobson measurement allows therefore to estimate the uncertainty of the Brewer adjusted Dobson TCO<sub>3</sub> values.

300 The statistics were obtained for various selected maximum ranges between the successive measurements, i.e. less than 1, 5, and 10 minutes. 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 the Dobson measurements with the uncertainty resulting from the observer's skill to perform the measurements. Table 2 presents the statistics of the differences between TCO<sub>3</sub> values obtained almost simultaneously for different data subsets.

305 The uncertainty of the Dobson observations for the subset with the nominal highest accuracy (DS&AD) is in the range of about [-1.15%, 1.09%] as derived from the [5<sup>th</sup>-95<sup>th</sup>] percentile range. 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 with nearly parallel DS measurements, not taking into account the specific cloud configuration and optical properties.

310 Table 2. The statistics of the differences between the Brewer adjusted Dobson TCO<sub>3</sub> values from two successive measurements taken at an interval of no longer than 10 minutes, for the period 1963-2019. The differences (in % of the mean of the two compared values) is shown for different sub-classes of the data and  $\mu$  ranges. n denotes number of the measurement pairs used in the calculations.

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

A few more changes are also necessary prior to the publication of the manuscript. Specific comments are provided below.

**In response to the reviewer's comments, we present the text with the suggested correction. The numbering of the lines applied to the revised manuscript (not with marked changes).**

*In the data files, or at least in an accompanying description file, please specify whether time is UTC or something else*

We add that UTC time was used (see l.346)

400 The results of all intraday measurements for the period 1963-2019 ~~have been~~ were previously stored at the PANGAEA data repository 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 ~~corrections described by~~ present analysis shows that the Brewer adjusted Dobson TCO<sub>3</sub> values are reliable for  $\mu < 4$

L7: please define that #84 is the serial number of the instrument.

Yes. This is done in accordance with the reviewer's comment (see l.7)

Abstract. The total column ozone (TCO<sub>3</sub>) measurements by the Dobson spectrophotometer #(serial No. 84) have been carried

L13: please add “which were” before “also performed”

Yes. This is done in accordance with the reviewer's comment (see 1.13).

values. To adjust this data to the Brewer spectrophotometer observations, **which were** also performed at Belsk, a procedure is

L20: Please explain that #1 and #2 are the serial numbers of the instruments

Yes. This is done in accordance with the reviewer's comment (see 1.21-22).

The monitoring of total column ozone (TCO<sub>3</sub>) started in 1924 in Oxford (the United Kingdom) with prototype of the Dobson instrument (DI). Before the Second World War, there existed data records from 2 stations Oxford (DI #serial No. 1) and Arosa (DI #serial No. 2) archived in the World Ozone and Ultraviolet Radiation Data Centre (~~WOUDEC~~). After the international

L22: “TCO” instead of “TOC” like in the rest of the document. At the same line, the authors probably mean that “the number of ozone observations increased sharply” instead of “The ozone observations were triggered”.

Yes. This is done in accordance with the reviewer's comment (see 1.23-24)

geophysical year in 1958, the total number of the stations with routine ~~POC<sub>3</sub>~~TCO<sub>3</sub> increased up to about 50. The **number of** ozone observations ~~were triggered~~increased sharply in the early 1980s ~~after following the~~ discovery of the ozone hole in the

L33: Delete “the”

Yes. This is done in accordance with the reviewer's comment (see 1. 34)

~~during~~improvements in the 1970s, it was available for ~~the~~ ozone monitoring in the early 1980s. However, differences between

L36: which ground-based network?

We explain this in the revised manuscript: “ in the TCO<sub>3</sub> ground-based observation network” (1.37-38)

1985), which replaced the previous ~~ones by~~Vigroux (1967);) coefficients, have been used operationally in the TCO<sub>3</sub> ground-based **observation** network since the early 1990s (Komhyr et al, 1993). Vanicek (2006) found that the difference could reach

L42: Similar results to those reported by Redondas et al. (2014), have been also reported by Fragkos et al., (2015).

Fragkos et al., (2015) paper has been added (see 1.43).

45 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 has not yet been recalculated ~~with use of~~using the

L47: In addition to Ball et al., the following study should be also cited: Steinbrecht, et al. (2017).

Steinbrecht et al. (2017) paper has been added (see 1.47)

~~stratospheric ozone in the NH midlatitudes~~,Steinbrecht et al., 2017; Ball et al., 2018). ~~Moreover~~,In addition, a surprising

L50: Please add references for the Arctic ozone depletion in 2020. For example:

Wohltmann et al. (2020); Manney et al. (2020); Inness et al. (2020).

Papers by Wohltmann et al. (2020) and Manney et al. (2020) have been added (see 1.53-54);

antarctic-ozone-hole-large-and-deep?). 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

L54: Delete “of the”

Yes. This is done in accordance with the reviewer's comment (see l. 56)

Poland, started monitoring ~~of the~~ atmospheric ozone (TCO<sub>3</sub> and the ozone vertical ~~profiles~~ profile by the Umkehr method) on

L55: Delete “including”

Yes. This is done in accordance with the reviewer's comment (see l. 57-58)

~~23-March 23, 1963. In Europe, there~~ There are only two stations in Europe with longer time series ~~including~~, Arosa (since 1926, Staehelin et al., 1998) and Hradec Kralove (since 1961, ~~Vaniček~~ Vanicek et al., 2012). The importance of the Belsk's

L65: “designed” instead of “deigned”

Yes. This is done in accordance with the reviewer's comment (see l. 67)

The Dobson spectrophotometer ~~is a double monochromator deigned~~ was designed to measure TCO<sub>3</sub> by the technique of the

Figure 2 and lines 90 – 95:

*First of all, the authors should explain how equations (1) and (2) were derived. Were all data shown in Figure 2 used to derive these equations?*

In the revised manuscript, we explain that the regression fit was used to part of the data with  $\mu$  in the range [2.8,4.0] (see l. 96-97).

types and their ~~configuration~~ positions in the sky. Therefore, the DS-ZS differences can sometimes exceed 5%. To eliminate ~~the~~ drift of the DS-ZS differences, the following transfer function is used ~~for large air masses: from the regression line fit to the relative differences between DS and ZS TCO<sub>3</sub> subsets for  $\mu \in [2.8, 4.0]$ ):~~

*Secondly, if the data shown in Figure 2 were used, then equation (2) has been calculated using a limited number of data points. Thus, I am not convinced that applying this relationship on future data would provide an accurate correction. Since data points for air mass above 4 are limited, and uncertainties in both the measurements of Dobson and MKII Brewer at such air masses are very large, I would recommend excluding data for air masses larger than 4 from the final, merged dataset.*

In the revised text we explain that the number of observations with  $\mu > 4$  was small and we decided to keep such data but should be treated with caution (see l.102-104)

115 is pertaining 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 TCO<sub>3</sub> observations were made at such high  $\mu$  values. The fixed correction of 1.015 is applied if  $\mu > 4.0$  but TCO<sub>3, ZS\*</sub> values should be treated with caution. The smoothed pattern of the DS-ZS differences (Fig.2b) is close to zero after the application

Line 129: Please define R/N

In the revised manuscript, we explain the meaning of R/N (l. 130-135).

calculation of the resulting R/N tables were ~~essential~~ of key importance for maintaining the quality of the 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 TCO<sub>3</sub> (e.g., Dobson, 1957).

Section 2.2: Adding a Table summarizing the campaigns (place, reference instrument, etc) would be useful.

Table 1 has been added. (see l. 147)

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

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

L172: Add reference(s) for the Brewer reference instrument. For example: Fioletov et al (2005).

Fioletov et al (2005) paper has been added (see l. 175)

(BS64) Mark II (~~with~~ single monochromator) ~~has been started~~ was launched at Belsk in 1991. Like other Brewers, the BS64 is a fully automated, self-testing, PC-controlled instrument ~~designed~~ 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

L180: “This ... spectrophotometers”. Please rephrase. The meaning of this sentence is not clear.

In the revised text, we explain that the difference concerns the Dobson and Brewer data (see l. 184-186).

of these TCO<sub>3</sub> values is small. i.e., standard deviation is less than 2.5 Dobson unit (DU). ~~This makes it difficult to directly compare the TCO<sub>3</sub> values with~~ The Dobson spectrophotometer provides instantaneous TCO<sub>3</sub> values, while the Brewer instrument gives the average of 5 observations, so this may be an additional source of differences between the spectrophotometers.

Figure 7: Even after the correction for the effective temperature there seems to be a trend in the ratio between the measurements from the two instruments (i.e., differences are ~+1% in 2002 – 2004 and ~-1% in 2018 - 2020). The authors should add some relative discussion (are these differences within the uncertainty of the merged dataset?).

However, the trend calculated in the Brewer-Dobson differences after the correction for the ozone absorption coefficients dependence to temperature is not statistically significant for the period 2002-2019. In the added text, we explain possible sources of the differences in the periods 2002-2004 and 2018-2020. (see l.217-220)

3% (ZS). The smoothed curved in Fig.7b provides that the Dobson TCO<sub>3</sub> values were ~1% lower (2002-2004) and ~1% higher (2018-2019) comparing with the Brewer values. Such discrepancies may be related to the Brewer ZS TCO<sub>3</sub> 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 result in overestimation (or underestimation) in relation to the Dobson TCO<sub>3</sub> values.

Section 2.3.3: Discussion about the effect of stray light can be also found in: Moeini et al. (2019)

In the revised manuscript, we discuss results by this paper. (see l.234-238)

2015). Moeimi et al. (2019) discussed the differences between TCO<sub>3</sub> 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<sub>3</sub> > 800 DU) was related to the instrument'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.

*In this latter paper the authors show that at very large ozone slant paths (i.e., for very large air masses) the role of stray light becomes exceptionally significant. That makes the measurements of both instruments unreliable. As I did earlier, I recommend again removing measurements for air masses larger than 4 from the analysis, as the uncertainties are already very large, solely due to the effect of stray light.*

We are aware of the stray light problem in the Brewer and Dobson spectrophotometers, so we propose the correction function for this effect. (1.300-303)

340 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 presence of stray light in spectrophotometers, causing underestimation of TCO<sub>3</sub> values at low solar elevations. The correction for the differences in the stray light effect in the spectrophotometers was proposed, see function (3), to reduce the B-D TCO<sub>3</sub> differences for low solar elevation. The stray light correction was not

In the conclusion section, we mention the following limitations on the use the merged time series (see l. 347-348)

2020). The ~~corrections described by~~ present analysis shows that the Brewer adjusted Dobson TCO<sub>3</sub> values are reliable for  $\mu < 4$  or slant TCO<sub>3</sub> up to 1400 DU (Fig.10).

*Ideally, the authors should correct the measurements of both instruments for the effect of stray light, which of course is not a trivial task. Instead, they have scaled the measurements of Dobson to the measurements of the Brewer at large air masses. Assuming that the scaling is perfect, stray light still affects the measurements of the Brewer, and subsequently the ozone series. In any case, the authors should discuss, and try to quantify, the uncertainties related to the stray light effect.*

True, the Brewer total column ozone was not corrected for the stray light effects. We propose the correction of the Dobson ozone to fit as close as possible the present Brewer data (without correction for the stray light). Figure 10 shows the Brewer adjusted Dobson column ozone is only slightly sensitive to the stray light. (l. 303-306)

340 proposed, see function (3), to reduce the B-D TCO<sub>3</sub> differences for low solar elevation. The stray light correction was not calculated separately for each instrument. However, Figure 10 shows, that the Brewer Adjusted TCO<sub>3</sub> values are only slightly sensitive to changes in slant TCO<sub>3</sub>, i.e., within the max-min range between 0.99 and 1.01 derived from the smoothed profile of the ratio between TCO<sub>3</sub> values non affected (slant TCO<sub>3</sub> < 800 DU) and affected by the stray light.

## **Response to reviewer 2 comments and suggestions.**

*This paper aims to provide a documentary of the long-term total ozone measurements at Belsk, Poland. This paper is well written and provides a great deal of details about record homogenization and calibration. I have studied stratospheric and tropospheric ozone variabilities for a long time, this manuscript fills me with some measurement history. In terms of data documentation, the material and presentation of the paper is nearly impeccable.*

*Whereas the authors point out the unexpected CFC emission and 2020 Antarctic ozone hole in the introduction, these issues are not discussed anywhere after the introduction. Since in the research community, the current mainstream seeks to address trends and variability attribution at detailed vertical structure/pressure surfaces, the total ozone measurements are rather handcuffed to answer the questions from a broader perspective. But the CFC emission and 2020 Antarctic ozone hole should be at least discussed further, for example, Belsk is a high latitude location, are the measurements affected by the Antarctic ozone hole in spring of 2020? As far as I recall, I have seen that the impact can be observed by Canadian ozonesonde records.*

In the introduction, we discussed the unexpected upward trend in CFC emissions in recent years, emphasizing the need to continue observing total ozone in the world.

This topic is not discussed later in the text, as manuscripts submitted to the journal should mainly focus on data description and procedures supporting the data quality. Therefore, the reviewer's suggestion to discuss the effect of the Antarctic ozone hole on total ozone in the mid-latitude NH using data from Belsk is left for further consideration.