

Response to Referee #1:

We thank referee #1 for their very helpful comments. Our responses are given below in black with the referee's comments in blue. The new text in the modified manuscript is given in red (italicized).

GENERAL COMMENTS

The manuscript by Zhao et al. analyzes the atmospheric conditions affecting the primary calibration of reference Brewers at four sites, with particular reference to the short-term variations of the total ozone column. A modelling framework is developed to simulate the effect of the ozone variations on the extraterrestrial calibration factor, using MERRA-2 reanalyses as input. The study aims at assessing: "(1) why Brewer primary calibration work can only be performed at certain sites... and (2) what is needed to assure the equivalence of calibration quality from different sites" (lines 104-106). While the answer to the first question is rather obvious, the second research question is very relevant to the ozone science. The paper is generally written in a clear way. Based on these considerations, I would recommend publication of the manuscript after some corrections.

We appreciate referee #1 for this very positive feedback.

SPECIFIC COMMENTS

- Structure. It may be a matter of taste, but I would recommend to follow a more traditional paper structure better highlighting the methods, the results and the discussion. I feel a bit confused, for example, when reading that Sect. 3 is titled "Primary calibration" and Sect. 3.2 is titled "Primary calibration method": what difference should the reader expect from the two sections?

We thank referee #1 point this out. The main difference is Sect. 3.1 only provides information on how to do one Langley plot, while Sect. 3.2 is about why and how to "combine" multiple Langley plots to achieve the calibration goal. Following the suggestion, we renamed the Sect. 3.2 from "Primary calibration method" to "ETC product".

- Different effects of short-term ozone variability. I am sure that the authors can introduce this topic in a more clear and tidy way (lines 320-330). Indeed, daily variations in ozone and instrumental factors can result in both random "noise" in the ETC determinations (appropriately tackled with type-A evaluation of the uncertainty) and systematic effects, notably in presence of recurring daily or sub-daily patterns (e.g., photochemistry? local pollution? instrumental artifacts?). I think that

this distinction requires further discussion, especially if the authors examine the linear and parabolic terms of the daily variations (e.g., how regularly these patterns occur?).

Following the suggestion, we provided more analysis of the linear and parabolic terms of the daily variations. The answer to this question is also related to the next one. So, we merged these replies together (i.e., see more detailed answers for the next question). We also want to point out there were comparisons made between Brewer #156 and QASUME (which was calibrated in the laboratory). Egli et al. (2022) shows that the difference between the lab calibrated approach (which is insensitive to systematic ozone variations that potentially affect the Langley-procedure) and the Langley-plot based approach is less than 1%.

An alternate approach has been demonstrated by Egli et al. (2022) using the QASUME spectroradiometer calibrated in the laboratory with SI-traceable radiation standards to retrieve the atmospheric TCO from direct spectral solar irradiance measurements without requiring an in-situ based Langley calibration, and thereby being insensitive to possible systematic ozone variations that potentially affect the Langley-plot based calibration. Collocated measurements between QASUME and Brewer #156 have shown good agreement in retrieved total column ozone with less than 1% difference.

- Relation between simulated and observed ETCs and reliability of MERRA-2 reanalysis. The authors state that:
 1. the "large day-to-day difference is mainly due to short-term ozone variability" (l. 297)
 2. they use MERRA-2 to "isolate the short-term ozone variability impacts" on ETCs (l. 360 - what does "isolate" mean, exactly?)

We have revised the sentence to make it clearer.

To avoid such instrument-related issues and meteorological factors, MERRA-2 reanalysis ozone data are used in an ETC simulation model developed to isolate the short-term ozone variability impacts (i.e., to exclude other instrumental and natural factors that could affect ETC) and also to avoid data gaps.

3. "the impact of linear and quadratic ozone variations can be independently assessed" (l. 405)
4. the uncertainty of MERRA-2 reanalysis is still too high to be used for correcting the single ETCs for short-term ozone changes (lines 600-603).

All things considered, a straight question is: is there a direct correlation between the ozone variations from MERRA-2 (e.g., in terms of the "b" or "c" coefficient) and the individual observed ETCs? Or should simulations using MERRA-2 data (and their good agreement with the observed behaviour, e.g. in Figs. 5e-h) only be interpreted as an average indication of the effect and its magnitude, depending on the site and the considered season? Can the authors further elaborate on the results reported at lines 406-408?

We thank the referee for this important question. So, we examined the agreement of MERRA-2 and Brewer observations in terms of their fitted daily 2nd-order polynomial lines ($TCO = a+b\Delta t+c\Delta t^2$). Figure R1 is an example of the results for Br#191, which shows the distribution of each fitting coefficient (blue bars are histograms of MERRA-2 fitting coefficients, while brown bars are histograms of Brewer #191's fitting coefficients). The values indicated in the legends are the mean \pm 1 sigma of the coefficients. Figure R1 shows that, statistically (on average), the ozone daily variations from MERRA-2 generally agreed with Brewer's observations (this is also supported by Fig. A3, which shows their % difference binned by the hour of local standard time), i.e., we did not see any strong evidence that MERRA-2 daily variation deviated from the observations.

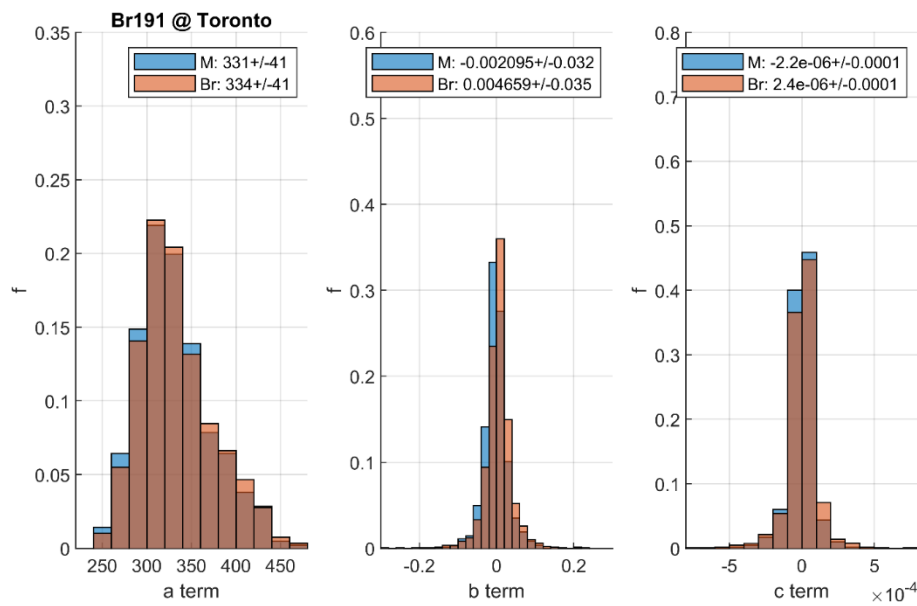


Figure R1. Histograms of 2nd-order polynomial fitting coefficients for daily ozone (for MERRA-2 and Brewer #191 at Toronto). The values indicated in the legends are the mean \pm 1 sigma of the coefficients.

As shown in the figure below (Fig. R2), we also examined the correlation between MERRA-2 and Brewer's fitted daily curves via their correlation coefficient (see blue bars). Here, we have >50% of cases the reanalysis and measurements show a strong correlation (>0.8). Note that for days with low ozone changes in a day (e.g., the ozone field is stable), the correlation between MERRA-2 and Brewer's fitted daily curve would be expected to have a relatively low correlation. Thus, if we further remove the days where ozone change in a day is less than median values, we have almost 80% of the dataset has $R^* > 0.8$. This means when we have moderate ozone daily changes (above median value), for about 80% of the time, MERRA-2 can represent >80% of ozone changes observed by Brewer in those days. Please note that, regarding the portion of the data that has been excluded in the R^* calculation (due to low ozone changes in a day), it also means both MERRA-2 and Brewer agree on those days that the ozone was relatively stable.

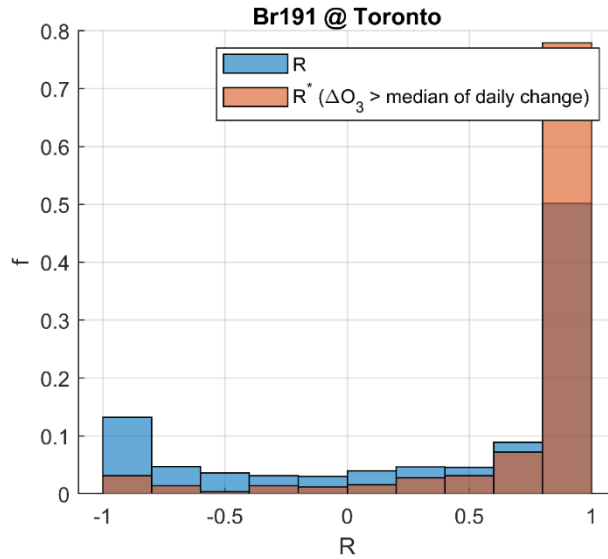


Figure R2. Histogram of correlation coefficients between MERRA-2 and Brewer observations. R is the correlation coefficient for the entire coincident dataset; R* is the correlation coefficient for the coincident data that has $\Delta O_3 \geq$ median of daily changes.

Further analysis of the differences between the fitted daily curves is shown below (Fig. R3). Here, for each day, the histograms of the difference between MERRA-2 and Brewer fitted 2nd polynomial curves are examined. The middle panel of Fig. R3 shows the standard deviation of the difference (MERRA-2 - Brewer), where the median value is only 3 DU. This result indicates that, for most conditions (in a given day), the difference between MERRA-2 and Brewer ozone is 3 DU on one sigma level. The distribution of the mean and median of the difference is not strongly deviated from the Gaussian shape (see the first and the last panel in Fig. R3). Thus, statistically, the MERRA-2 ozone followed a similar pattern as observed ozone (with some minor offset for this site (Toronto), which was also reported in our other MERRA-2 vs. Brewer analysis in Appendix A).

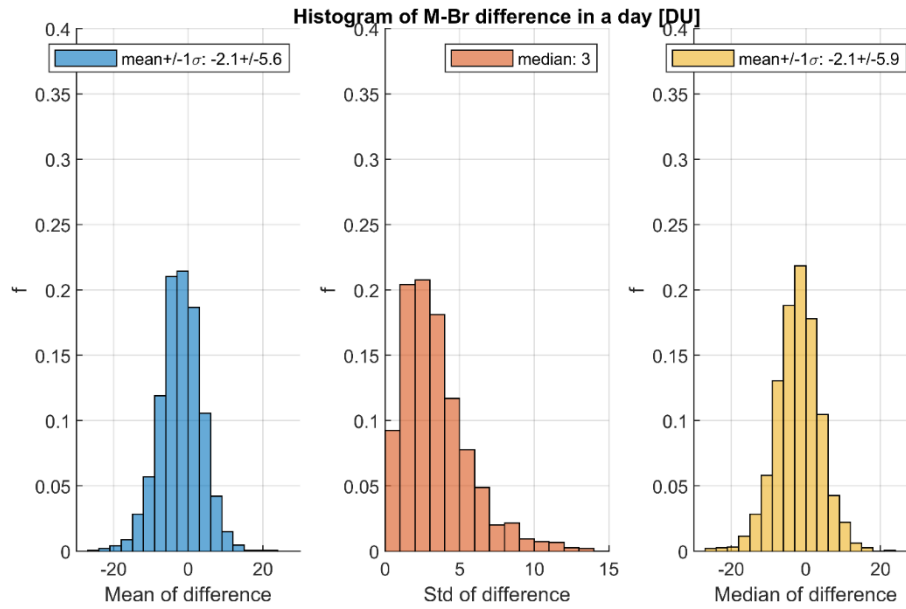


Figure R3. Histogram of the difference between MERRA-2 and Brewer fitted ozone curves.

Figures R4–6 show the same analysis as Fig. R1, but for all 11 Brewers included in this work. Figure R4 is the summary of fitted constant term for all 11 instruments; Fig. R5 is the summary of fitted linear term, and Fig. R6 is the one of fitted quadratic term.

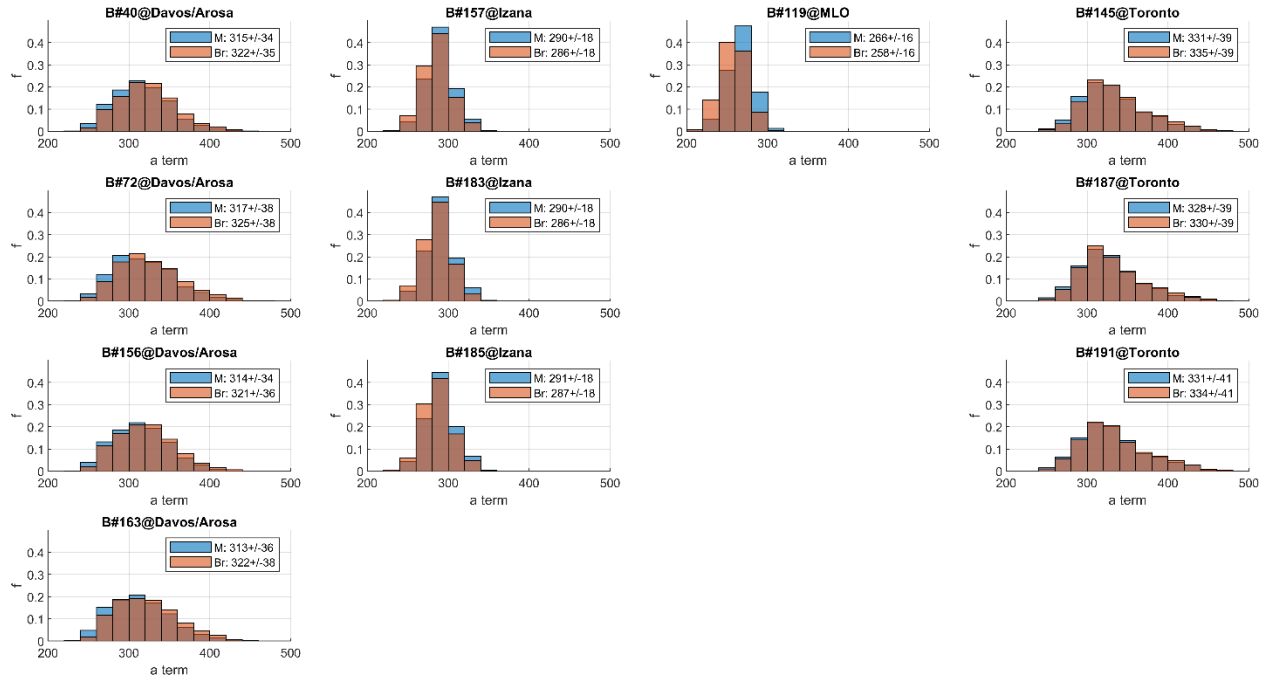


Figure R4. Histograms of 2nd order polynomial fitting coefficient (0 order term; constant term) for daily ozone. The values indicated in the legends are the mean \pm 1 sigma of the coefficient.

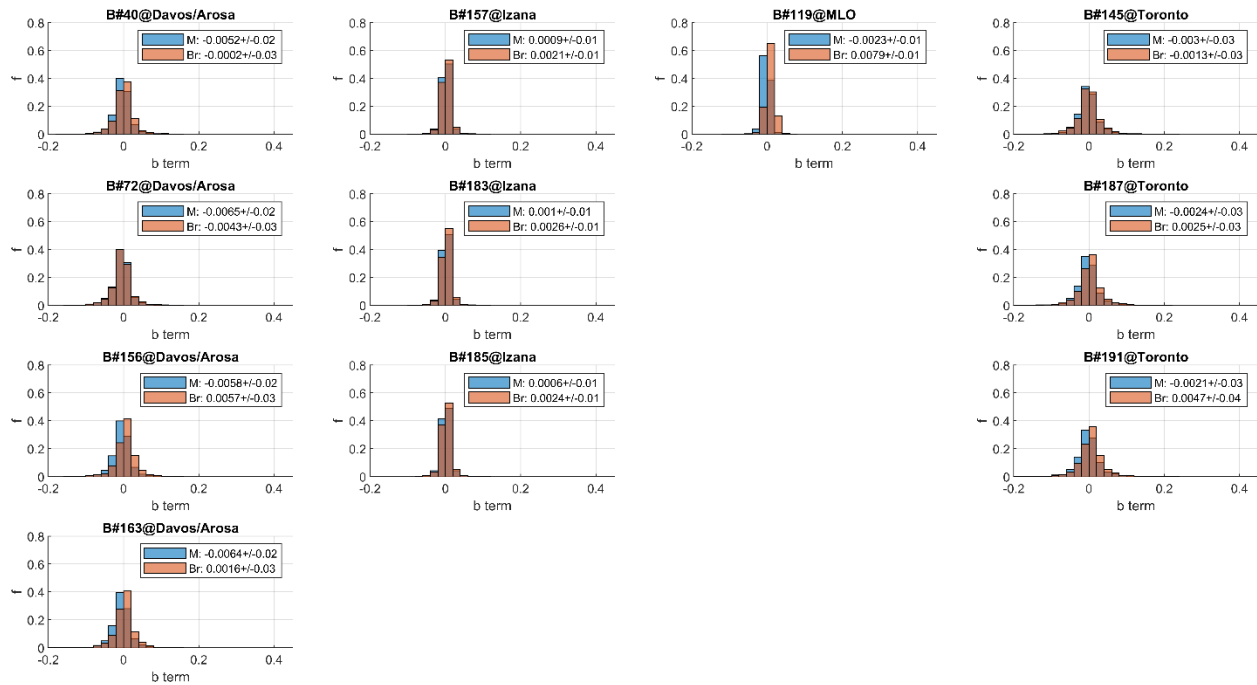


Figure R5. Histograms of 2nd order polynomial fitting coefficient (1st order term; linear term) for daily ozone. The values indicated in the legends are the mean \pm 1 sigma of the coefficient.

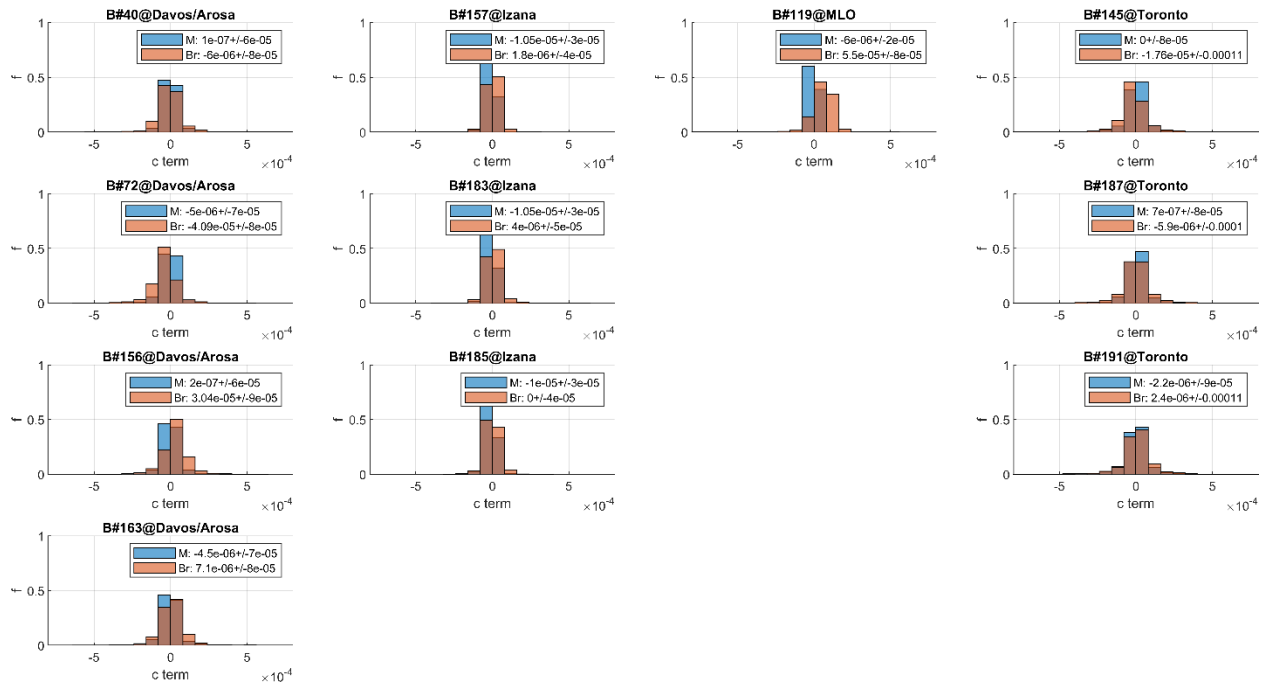


Figure R6. Histograms of 2nd order polynomial fitting coefficient (2nd order term; quadratic term) for daily ozone. The values indicated in the legends are the mean \pm 1 sigma of the coefficient.

However, we admit that the agreement between reanalysis and observation is not perfect. There are many factors that could contribute to the difference, which include but are not limited to, such as model resolution, line-of-sight of Brewer instrument, vertical sensitivity difference, etc. However, MERRA-2 is good enough for the current modelling work to prove that ozone variability in the short-term is the major challenge in Brewer primary calibration work. Figures R4–6 have been added to Appendix A as Figs. A4–6, and some of these discussions are included in the manuscript (Sections 3.3, 6, and the Appendix).

Simple TCO variation amplitudes (i.e., maximum value subtract minimum value for that given period) for whole daytime, a.m., and p.m. sessions are reported in Fig. 3d to f, with the probability of variation amplitudes less than 1.5 DU (suitable conditions for Brewer Langley calibration work) shown in the legends. For example, the results (Fig. 3e) show that Brewers at MLO and Izaña have a better chance (e.g., 55% and 45% for morning sessions, respectively) to have a good stable short-term ozone field to produce high-quality individual ETCs via Langley plot techniques than the ones at Davos (23%) or Toronto (20%). Statistics of MERRA-2 and Brewer's fitted daily ozone variations are provided in Appendix A (see Figs. A4–6).

Currently, other ground-based instruments, satellites, or reanalysis models (such as MERRA-2 used here) do not have sufficient accuracy and precision to support Brewer's primary calibration work. In general, MERRA-2 can capture the general pattern of ozone daily changes. The median value of the standard deviation of the difference between MERRA-2 and Brewer ozone is only 3 DU (about 1%). However, limited by many factors (such as spatial and temporal model resolution, instrument line-of-sight, etc.), currently, the modelled ozone could not perfectly reproduce the observations record.

These small differences in seasonal and diurnal patterns should only have a limited impact on the modelled daily ozone variation pattern or the ETC simulation model. This is also confirmed by the acceptable agreement found between the model and observation-based calibration site condition results (e.g., see Fig. 7). We also examined the agreement of MERRA-2 and Brewer observations in terms of their fitted daily 2nd-order polynomial lines ($TCO = a + b\Delta t + c\Delta t^2$). The histograms of their fitted coefficients are shown in Figs. A4–6. Statistically, MERRA-2 can follow the local daily ozone variation pattern (the fitting terms by using MERRA-2 and Brewer observations are agreed on a 1-sigma level).

- Sect. 4: is SO₂ an interfering factor at MLO?

We thank the referee to point out this special condition factor for the MLO site. Yes, theoretically, SO₂ could affect the calibration of Brewer. However, we also must point out that the wavelengths of Brewer's operational ozone algorithm (nominal values at 310.1, 313.5, 316.8, and 320 nm; the four slits with the longest wavelengths measured by Brewer) were selected to be located at wavelengths that eliminate differential absorption due to SO₂ (Kerr, 2010). Figure 6 of Evans et al. (1981) shows measurements of large values of SO₂ (>~50 DU) during the passage of volcanic debris from Mount St. Helen which erupted in late May, 1980. These measurements were made with the Brewer Mark I version. There is no suggestion that SO₂ impacts the ozone measurement, although it is difficult to tell since ozone itself is changing. Figure 4 of Kerr et al., (1985) shows another major SO₂ pollution event (~40 DU) in August, 1983 observed by

two Mark II instruments (#5 and #8). In this case both instruments show no significant impact by SO₂ on the ozone measurements which remained relatively stable during this event.

In addition, the observatory (MLO) is 2000 m above the volcano, so degassing SO₂ from Kilauea volcano typically cannot reach the observatory. In fact, for the world reference instruments, we rarely detected any SO₂ signal during the previous calibration trips also because typically prevailing winds (from east to west) direct SO₂ plumes away from the observatory. Figure R7 is the histogram of SO₂ column measured by Brewer #119 at MLO. As Fig. 7 shows, SO₂ at MLO is typically within the noise level, and even 5 DU SO₂ is very rare for MLO.

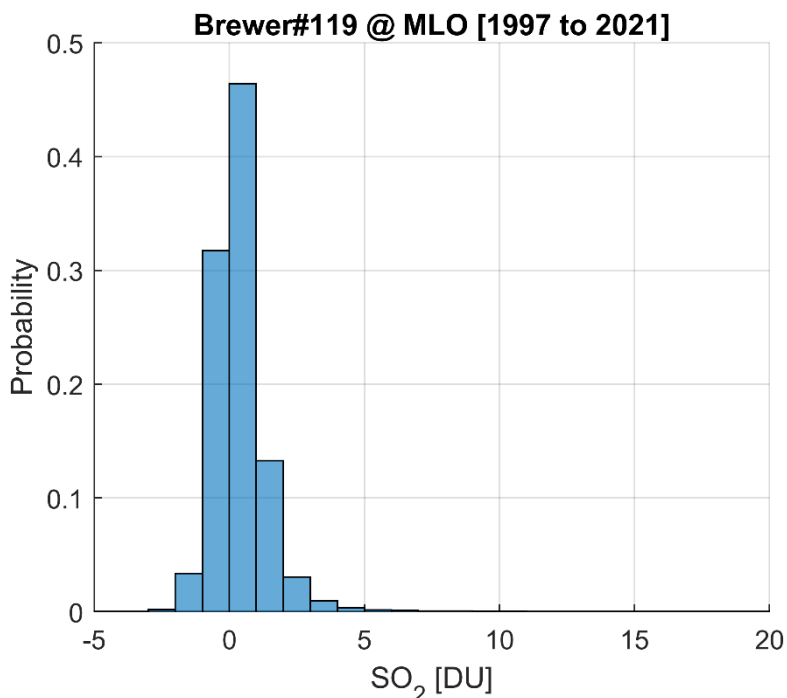
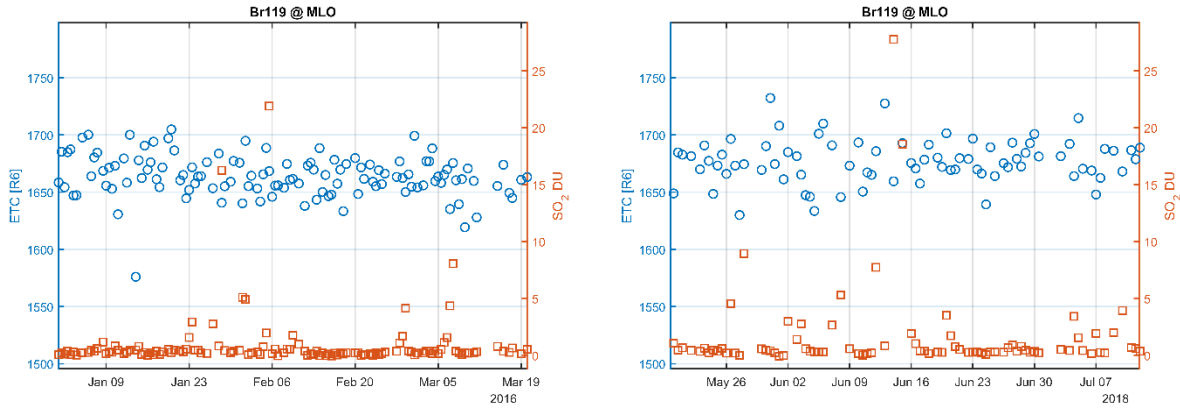


Figure R7. Histogram of column SO₂ measured by Brewer #119 at MLO.

In 2018, the Kilauea volcano eruption occurred and was still moderately active in 2019 while the ECCC Brewer team was there doing calibrations. When the winds became more southerly, the instrument actually saw SO₂ values reach as high as 10 DU. Such data are in fact good for the ETCSO₂ calculation. To further demonstrate if presence of high SO₂ could impact ETCO₃, we also plotted the fitted ETCs along with the high SO₂ observed for 2016 and 2018 cases (note these are rare cases, and were not the periods that Brewer world reference instruments were performing calibration at MLO). Figure R8 shows that, even when SO₂ column > 20 DU, there is no indication that the fitted ETCO₃ values are driven by the SO₂ values.



This information has been included in the revised manuscript (Sect. 4.2 and Appendix B).

Meanwhile, due to the different characteristics of each site, different optimized strategies must be selected by the AEMET and ECCC Brewer teams in practice. For example, it was found the MLO site has more cloudy conditions during afternoon sessions and thus, more observations must be made to ensure a good balance of morning and afternoon sessions. *Another factor that might impact primary calibration at the MLO site is the SO₂ plume from the volcano. However, we must point out that the wavelengths of the Brewer operational ozone algorithm were selected to be located at wavelengths that eliminate differential absorption due to SO₂ (Kerr, 2010). Previous work shows no evidence that SO₂ will affect Brewer ozone observations (e.g., Evans et al., 1981; Kerr et al., 1985). Examples of fitted ETC values with observed SO₂ are shown in Appendix B.*

The MLO is 2000 m above the Kilauea volcano, and thus degassing SO₂ from the volcano typically cannot reach the observatory. In addition, with the world reference instruments, we rarely detected any SO₂ signal during previous calibration trips because typically prevailing winds (from east to west) direct SO₂ plumes away from the observatory. Figure B2 is the histogram of SO₂ column measured by Brewer #119 at MLO. As Fig. B2 shows, SO₂ at MLO is typically within the noise level, and even 5 DU SO₂ is very rare for MLO. The example timeseries from 2016 and 2018 demonstrated that the fitted ETC values are not driven by the variation of SO₂, even when SO₂ column values > 20 DU.

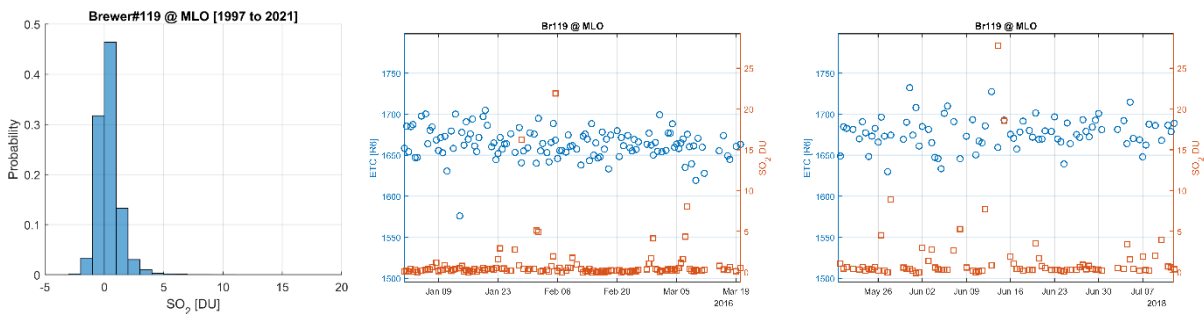


Figure B2. (left) Histogram of column SO₂ measured by Brewer #119 at MLO (middle and right). Example timeseries of fitted ETCs and observed SO₂.

TECNICAL REMARKS

- Title: I'm not a native English speaker, but "site-specified" sounds a bit odd (should it be "site-specific"?)

Done.

- 1. 29, "stable short-term ozone field": "stable ozone field in the short term"

Done.

- 1. 36: notice that if R6 units are divided by the differential absorption coefficient, the same quantity can be expressed in DU (relative to airmass 1). The equivalent quantity in DU can thus be specified in the text for greater clarity

Thanks for this good suggestion. However, the differential absorption coefficient ($\Delta\alpha$) for each Brewer is different. The value is determined by wavelengths and resolution of an individual instrument, and also the ozone cross sections (and the effective temperature) that have been selected (e.g., such as using BOp or IUP cross sections) (e.g., Redondas et al., 2014; Gröbner et al., 2021). Thus, if we select a "typical" $\Delta\alpha$ (e.g., 0.34) and use airmass = 2, we can "convert" R6 into DU. For example, 5 R6 units is equal to 1.47 DU (with $\mu = 1$ and $\Delta\alpha = 0.34$). Such conversion provides a clear meaning of the value, however, it will be more difficult for us to quantify the true instrument response alone. As the main idea of this work is to set up and examine the technical standard for Brewer calibration work (e.g., primary calibration to meet the 5 R6 unit goal), we would prefer to continue to use the R6 unit. We have included such information in the revised work to make this clearer for readers.

For a Brewer instrument, the goal of such calibration is to derive its unique ETC value (that can be used in TCO calculation), with uncertainty within ± 5 R6 units (Zhao et al., 2021). Here R6 is a measurement-derived double ratio in the actual Brewer processing algorithm, corresponding to the measured slant column ozone (e.g., Savastouk, 2006; Zhao et al., 2021). Note that in typical conditions (e.g., $\Delta\alpha = 0.34$ and $\mu = 2$), 5 R6 units is equal to 0.74 DU or about 0.25% for a typical ozone value of 300 DU.

- 1. 117: it should be mentioned that the second "channel" is used for SO₂ retrievals

Done.

The Brewer spectrophotometer is a modified Ebert grating spectrometer that was designed to measure almost simultaneously the intensity of radiation at six UV channels (nominal wavelength at 303.2, 306.3, 310.1, 313.5, 316.8, and 320.1 nm). The first channel is almost exclusively used for wavelength calibration, *the second channel is used for SO₂ retrieval*.

- 1. 124-125: this formula should be written in a distinct line, so that the definition of F can be referenced more easily if needed (e.g., line 225)

Done.

F , $\Delta\alpha$, and $\Delta\beta$ are the linear combinations of the logarithms of the measured intensity, the effective ozone absorption and the Rayleigh scattering coefficients, respectively. For example,

$$F = -\log_{10}(I_3) + 0.5 \log_{10}(I_4) + 2.2 \log_{10}(I_5) - 1.7 \log_{10}(I_6), \quad (2)$$

where I_3 to I_6 are the photon count rates at the last four longer wavelength channels (Kerr et al., 1985).

With assumptions that TCO values (Ω) are constant through the calibration session (half-day) and aerosol has negligible impact, the instrument response F_i (see Eqn. 2) adjusted for instrumental (dead time, dark counts) and some atmospheric (Rayleigh scattering) factors is a linear function of air mass (μ_i):

$$F_i = ETC + 10\Omega\Delta\alpha\mu_i + e_i \quad (3)$$

where, i is the observation number, $\Delta\alpha$ is the effective ozone absorption, and 10 is a scaling factor used in the Brewer software, ETC is the extraterrestrial constant (here, $ETC = -10^4 \times F_0$).

- 1. 125-126, "the last four longer" --> "the four longest"

Done.

- 1. 146: notice that Cede et al., 2006 employ MkIII Brewer, which clashes with the premise "NO₂, by Mark IV only"

Done.

The Brewer spectrophotometer provides data products that include column ozone (e.g., Kerr, 2002; Kerr et al., 1981), column sulphur dioxide (SO₂; e.g., Fioletov et al., 1998; Zerefos et al., 2017), column nitrogen dioxide (NO₂, *by Mark III and IV*; e.g., Kerr et al., 1988; Cede et al., 2006; Diémoz et al., 2021), spectral UV radiation (e.g., Bais et al., 1996; Fioletov et al., 2002), aerosol optical depth (AOD) (e.g., Kazadzis et al., 2005; Marenco et al., 2002; Diémoz et al., 2016; López-Solano et al., 2018), and effective ozone layer temperature (Kerr, 2002).

- 1. 168: "wavelength" --> "wavelengths"

Done.

- 1. 191-195: isn't it a repetition of what is already said in the Introduction?

We removed the repeated part.

Four long-term Brewer calibration and/or operation sites are included in this work: Arosa/Davos, Switzerland; Izaña, Spain; MLO, Hawaii, U.S.A; and Toronto, Canada (see Table 1 for details). ~~MLO and Izaña are the calibration sites for the world reference triads and the European regional triad, respectively. Notably, there are four Brewer triads in operations worldwide, the Swiss Brewer triad (Stübi et al., 2017), the European regional Brewer reference triad (León Luis et al., 2018), and two world Brewer reference triads (Fioletov et al., 2005; Zhao et al., 2021). Arosa/Davos, Izaña, and Toronto are the operation sites for the Swiss triad, RBCC-E, and the world references, respectively.~~

- Eq. (2): notice that the equation is different from (1). In addition to the 10 factor, the sign of the ozone term is opposite. Please, use only one convention for the sign of the differential coefficient (e.g., define it positive). Also, use either the "F0" or "ETC" expression

We thank the referee to point out this issue, and we apologize for the confusion. The main difference between Eqn. (1) and Eqn. (2) (in the revised paper, corresponding to Eqn. (3)) is the definition of instrument response. In the original definition (e.g., Kerr, 2010), the instrument response (F) is a negative number; for convenience (and due to the storage limit in old days), the intensities (I) measured by the instrument were stored in $10^4 \log_{10}(I)$ scale. The ETC is converted to a positive number via $ETC = -10^4 \times F_0$ (it was provided in Line 231 in the original manuscript). The factor of 10 in Eqn. (2) was for the fact that the Brewer algorithm has this -10^4 factor included and also we converted the total ozone from the unit of cm to the Dobson Unit ($1 \text{ DU} = 10^{-3} \text{ cm}$). To make the equations here to be more consistent and clearer, we modified the Eqns. 1–2, and the conversion of ETC from F_0 accordingly (i.e., re-defined the F values to be positive). We also provided a simple explanation of factor 10 in the text.

The four longer wavelengths are used for the TCO (Ω) retrieval via the following equation:

$$F - \Delta\beta \cdot m = F_0 + \Delta\alpha \cdot \Omega \cdot \mu \quad (1)$$

where, m and μ are the enhancement factors for the slant-to-vertical path length of the direct radiation for air and the ozone layer respectively (also known as the air mass factors).

With assumptions that TCO values (Ω) are constant through the calibration session (half-day) and aerosol has negligible impact, the instrument response F_i (see Eqn. 2) adjusted for instrumental (dead time, dark counts) and some atmospheric (Rayleigh scattering) factors is a linear function of airmass (μ_i):

$$F_i = ETC + 10\Omega\Delta\alpha\mu_i + e_i \quad (3)$$

where, i is the observation number, $\Delta\alpha$ is the effective ozone absorption, and I_0 is a scaling factor used in the Brewer software (this factor is to accommodate that the measured intensities are stored in $10^4 \log_{10}(I)$ scale and to convert the ozone column unit from cm to the DU), ETC is the extraterrestrial constant (here, $ETC = 10^4 \times F_0$).

- 1.2

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