EUBREWNET RBCC-E Huelva 2015 Ozone Brewer Intercomparison

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Response to Reviewer #2

Comment: The paper "EUBREWNET RBCC-E Huelva 2015 Ozone Brewer Intercomparison" by Redondas et al. describes some of the main findings from an international comparison campaign of Brewer spectrophotometers. After an introduction about the Brewer ozone retrieval algorithm and the calibration transfer techniques, particular attention is given to an empirical parametrisation/correction of stray light applied to the single-monochromator instruments. A short discussion about the "standard lamp correction" to track the radiometric stability of the spectrophotometers is also provided.

In my opinion, the paper potentially raises the following important questions:

1. What is the maximum attainable reproducibility by well-calibrated Brewer spectrophotometers?.

2. What are the most common sources of error/instability in the Brewer measurements? How can they be identified and solved during an intercomparison?.

3. How important is the stray light effect on ozone estimates and what techniques can be used to overcome this issue?.

4. How good is the agreement among reference instruments used to calibrate the Brewer network?.

Therefore, the manuscript, in principle, addresses relevant scientific questions within the scope of ACP. However, I have two main concerns related to the paper:

1. The stray light and the standard lamp corrections should be discussed more properly (cf. Specific comments);
2. The manuscript resembles more to a technical report than a scientific article (especially considering that the manuscript has been submitted to ACP).

The previously listed scientific questions (1-4) deserve a deeper discussion, and should be better enhanced (e.g., they should be presented in the introduction, together with a set of bibliographic references, and answered in the main text through quantitative results). Technical details (e.g., determination of the dead time, dispersion function, etc.) that would be suitable for a report should be omitted in the present paper if not relevant to the scientific discussion. A reorganization of the paper, keeping theory and results better apart, would improve readability (cf. Technical corrections).

Once these remarks are properly addressed, the paper can be published in ACP.

Answer:

We have reorganised the paper to address these main questions. The stray light and the standard lamp corrections are discussed on the Material and Methods section and the introduction included reference on the main topics.

In this work we focus on the reproducibility of the EUBREWNET network, the precision and accuracy of well maintained triads which serve as references of the Brewer ozone measurements are described in other studies (Fioletov et al., 2005; Stübi et al., 2017; León-Luis et al., 2018) and we think that this is outside the topic of this work.

We add a table of the comparison of the reference instruments during the RBCC-E campaigns and the corresponding calibration report. The agreement is generally around +/- 0.5% but with exceptions. In contrast to the RBCC-E travelling reference who is transported by boat/car to Huelva and as hand luggage using two extra-seats of the plane, to Arosa campaigns, the others travelling references (IOS #017 and Kipp&Zonen #158) are usually transported by cargo and can have issues during transportation that are reflected in the table. Also, due to the instrumental changes for example #158 has a new PMT and new electronics during Arosa 2014 and the SL do not reflect this change.

Specific comments

Comment: It is rather trivial that the comparison between single- and double-monochromator instruments improves when a stray light empirical correction, obtained from the comparison itself, is applied back to the same set of data. What is not obvious, in my opinion, is that the correction obtained during the intercomparison can be also used to improve accuracy when the Brewer is moved back to the home institution after the campaign. This would be an important conclusion, but some points should be addressed:

Table 1. Reference Comparison during RBCC-E campaigns

Location	year	#017	#158	#145	Report
Arosa	2008	-0.6			(Redondas and Rodriguez-Franco, 2008)
Huelva	2009	-0.6	0.8	-0.1	(Roozendael et al., 2012)
Arosa	2010	-0.6			(Roozendael et al., 2013b)
Huelva	2011	-0.1	-0.2	-0.6	(Roozendael et al., 2013a)
Arosa	2012		-0.1		(Redondas et al., 2015)
Huelva	2013	-1.0	0.7		(Redondas and Rodriguez-Franco, 2015a)
Izaña	2014			-2.2	(Redondas et al., 2014)
Arosa	2014	-1.2	1.5		(Redondas and Rodriguez-Franco, 2015b)
Huelva	2015	-0.5	-0.5		this work

1. it should be proved (or discussed) that the correction only depends on the instrumental characteristics and not on the measurement site;

2. stray light should be characterised, during the intercomparison, for the full range of slant ozone values reached during normal operation. This is particularly important for single-monochromator instruments located at high-latitude stations. Can the authors state that the OSC range during the intercomparison is wide enough?

3. the authors affirm that "These parameters, determined in several campaigns, have been found to be stable" (page 6 l. 8). This is a key point: can they show some quantitative data demonstrating that the correction is stable over time?

Comment:

Answer: We agree with the suggestion and we think we can prove our affirmations. As described in the introduction, one of the regular RBCC-E campaigns were the Nordic campaigns, within these campaigns the FMI MKII Brewer #037 operating at Sodankyla since 1988 was calibrated four times, at Izana in 2009, 2011, and 2015 and at Sodankyla (Finland) in 2011 (Roozendael et al., 2013b, 2014).

From these measurement campaigns, we found

- The stray light correction obtained during the first campaign where applied to the subsequent campaigns obtained a very good agreement, better than 0.5% on the 300-1800 range.

- The correction is valid also when the spectral response of the instrument is changed, we detect a change on the ETC during the last campaign (2013), and the application of the new ETC with the 2009 stray parameters give also very good results.

- The stray light parameters (k and s) obtained during different campaigns (Table 2) are in agreement when we consider the confidence interval of the adjustment.

- During the calibrations at Huelva, the measurement schedule is carefully defined to maximize the observations at high airmass. As also described in the campaign conditions Section around 30% of the simultaneous observations are performed

Table 2. Summary of the FMI #037 from (Roozendael et al., 2014) calibration constants including the stray light parameters k and s, the intercept F0 and the ETC constant calculated using the standard 1-parameter method (F01P) and the standard lamp R6 ratio reference value (R6ref). R2 is the coefficient of determination for the power-law fitting.

Campaign	k	k(95%CI)	s	s(95%CI)	F_0	F_0(95%CI)	R2	F01P	R6Ref
Izo2009	-12	[-17.58,-6.41]	4.79	[3.79,5.78]	3117	[3112,3123]	0.942	3115	1880
Sdk2011	-12.66	[-18.65,-6.67]	4.56	[3.88,5.23]	3104	[3091,3118]	0.99	3115	1880
Izo2011	-18.29	[-24.51,-12.07]	3.97	[3.19,4.76]	3106	[3102,3111]	0.987	3115	1880
Izo2013	-11.37	[-17.50,-5.25]	5.54	[4.42,6.66]	3119	[3112,3126]	0.986	3120	1870

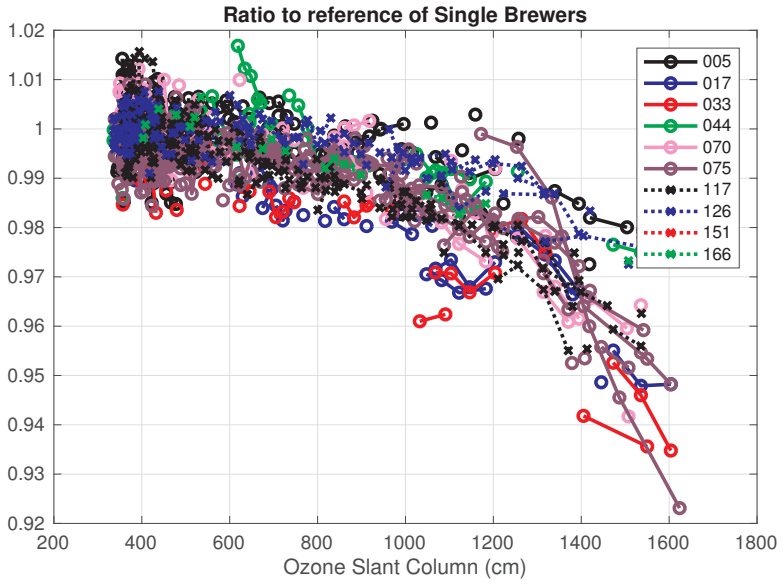

Figure 1. Ratio to reference on Single Brewer, the stray light free region is generally up to 800 DU were underestimation start and is determined for each instrument based on the one-parametner/two-parameter calibration agreement

with OSC> 600, 15% of that >900 reaching the 1600 DU. During these campaigns at Izaña, we can get 1800 DU reaching 2000 at Sodankyla.

- Figure 1 shows the ratio of the single brewer participating in the campaign, here we can see that the OSC free region which we use for calibration start for some instruments at 600DU and is almost evident at 1000DU, utilizing observations up to 1600 DU we have enough measurements to determine the stray light constants.

Comment: The section about the SL correction (page 9) is quite inconclusive. It is demonstrated that the SL correction does not improve the accuracy for some Brewers (while it does for others) and that the only way to verify it is an intercomparison against a reference instrument. In that case, how should the Brewer data be reprocessed from one intercomparison to the

next? My concern is not to spread the idea that the Brewer data quality is aleatory and that the community does not know how to reprocess the data for improving their quality.

Answer: We try to rephrase this but this is exactly the fact, the Brewer calibration is tracked by the SL measurements, without any external comparison we are not able to determine if the internal lamp properly tracks the changes of the spectral response of the instrument or not. This is the main reason why a regular calibration of the instrument is needed (every two years) and why the final data in EUBREWNET (Level 2.0) is achieved after the calibration. Of course stable instruments have also stable SL record and maintain their calibration and no reprocessing is required, for our experience small changes are tracked very well with the standard lamp but this does not always happen with huge changes due mostly to major issues in the instrument.

Technical corrections

Comment: page 1 l. 4-6: omit the reference to UV and QASUME in the abstract if the UV results are not discussed in the text;

Answer: removed

Comment: page 1 l. 4: "Twenty-one". It could be useful to mention already in the abstract how many single- and double-monochromator instruments have been studied;

Answer: Added the number of doubles and singles, also this information is added to the table. The proportion of single and double Brewer in the campaign is approximately the same as the network

Comment: page 1 l. 7: at the first occurrence, use "spectral stray light" instead of only "stray light", to distinguish it from other sources of stray light (e.g., multiple scatter stray light n the field of view);

Answer:

Added a description of the stray light as was also requested by Referee #1

Comment: page 1 l. 9-10: omit 76% and 50% percentages (16/21 is easy to calculate, and 10/21 is 47%, not 50%);

Answer: done. For the statistics we do not count the instrument #151 as it is not operative and is not providing observations.

Comment: page 1 l. 10: state the air mass range relative to the 1% and 0.5% thresholds;

Answer: included the ozone slant column rather than the airmass range, as the stray light effect depend on this.

Comment: page 1 l. 11: enhance the outcomes of the paper, e.g. why / to whom those findings are important?

Answer: added, see general comments.

Comment: page 2, Sect. 1: I would expect a more sound introduction, focusing on scientific issues (and related literature, e.g. on the stray light effect, etc.), rather than a summary of the previous campaigns. Also, since the previous technical reports

are mentioned, it should be specified what the novelty of the present study is;

Answer: Each RBCC-E campaign advances our understanding of the Brewer calibrations in general and specifically what areas still require attention. In this 2015 campaign we have introduced a formal approach to characterization of the internal instrumental stray-light , the filter attenuation correction and the algorithm for correcting for its effects on the TOC calculations, and these improvements were introduced on the EUBREWNET calculation.

Comment: page 2 l. 17: "Figure" instead of "Fig." at the be- ginning of a sentence

Answer: done .

Comment: page 3 l. 2: table 2 is cited before table 1, please reverse the order of tables;

Answer: done

Comment: page 3 l. 3: the measurement site should be better described, since the local characteristics of the measurement site impact of the results of the campaign;

Answer: A description of the site and the conditions of the intercomparison has been added.

Comment: page 3 l. 4-5: since UV radiation is out of the scope of this paper, omit the references to QASUME and WRC-UV;

Answer: The UV radiation is not covered by the paper but is an important aspect of the campaign. Moreover some of the aspect of the calibration are cross related like the wavelength calibration.

Comment: page 3 l. 8-21: those paragraphs have nothing to do with Sect. 1.1, entitled "The X RBCC-E campaign". Omit them and reference the EUBREWNET paper (https://www.atmos-chem-phys-discuss.net/acp-2017-1207/) in your Introduction;

Answer: done

Comment: page 4 Eq. 1: if alpha is defined as in Eq. 3, Eq. 1 should read (ETC-F)/(alpha*mu) instead of (F-ETC)/(alpha*mu). Otherwise, if alpha is defined negative (as in the Brewer literature), Eq. 3 should have a minus sign;

Answer: corrected

Comment: page 4 l. 8: define what "double ratios" are, for the unexperienced reader. "corrected for the Rayleigh effects" -> "to which the effect of Rayleigh scattering has been subtracted (Eq. 2)";

Answer: corrected in text.

Comment: page 4 l. 16: "verify" -> "satisfy";

Answer: done

Comment: page 4 Eq. 5: Eq. 6 is explained in the following text, please explain Eq. 5 as well;

Answer: added an explanation and reference

Comment: page 5 l. 1: "calibration" -> do you rather mean "characterization"?

Answer: corrected

Comment: page 5 l. 2: "filter attenuation" -> has any filter already been mentioned in the text?

Answer: A short description of the instrument is added.

Comment: page 5 l. 3: "The wavelength calibration allows to..." -> "An accurate determination of the operational wavelengths is needed to". "are" -> "is";

Answer: The paragraph has been modified following your suggestion

Comment: page 5 l. 6: "Finally, the ETC transfer is performed by comparison..." -> "Finally, the ETC must be determined by transfer from a reference Brewer (Sect. 2.1) or, in the case of the reference instruments, by the Langley method" (include bibliographic references about the Langley method);

Answer: The sentence has been rephrased and some references to the Langley method were added.

Comment: page 5 l. 8-9: "changes ... wavelength calibration will affect the final ETC and changes in the wavelength calibration will affect also the final ETC" -> is there any difference between the two sentences?

Answer: corrected

Comment: page 5 l. 18: "calibration" -> "radiometric";

Answer: The word "radiometric" suggests an absolute calibration of the instrument. However, the Brewers present a relative calibration, so we think that it is more correct to use the word "calibration" for this article.

Comment: page 5 l. 20: "for simultaneous measurements BY BOTH INSTRUMENTS";

Answer: corrected

Comment: page 5 Eq. 7: explain that "i" does not refer to wavelength, as in Eqs. 2-6, but to the sample (better: use a different subscript). Correct the sign "-" to "+" according to Eq. 2 (cf. my previous comment);

Answer: corrected

Comment: page 5 l. 27: define the "stray-light free region" from a quantitative point of view;

Answer: A detailed definition of the stray-light free region is defined, from a quantitative point of view this could be defined as the region where the underestimation of the ozone is less than 0.3% but it is difficult to address because this difference will also depend on the calibration constants of the instrument. To avoid this the agreement of one point calibration with the two point calibration is used to determine the stray light free region for every instrument, which range from 900 DU to 600 DU.

Comment: page 6 Fig. 3: Figs. 3 and 4 are very similar. Consider keeping only one of them. Add units of ozone slant path (cm STP);

Answer: The figure 3 is the standard operating procedure where as the Figure 4 shows the stray light calculation, as the data are the same we try to use only Figure 4 but then the standard operating procedure is not clear, so we think that is worthwhile to maintain the two, figures.

Comment: page 6 Eq. 8: the "F0" notation of Eq. 4 may be confused with "Fo" of Eq. 8. Consider using a different notation;

Answer: corrected

Comment: page 6 Eq. 8 and 9: as far as I understand, the k(X*mu)^2 term can be attributed to either the F's or to the ETC's, but not to both of them at the same time. In this case, the text should be clearer;

Answer: Mathematically it can be considered to both terms, it is a correction to the measured counts but as is determined with the ETC, and the correction is similar to to the Standard Lamp correction. This is corrected in the text.

Comment: page 6 line 7: "stray-light free OSC region" -> this expression is misleading, since the k(X*mu)^s cannot be zero for mu>=1 (or reformulate the correction term to be zero for mu=1). Also, could the ETC be directly retrieved from the fit, together with k and s?

Answer:

The non-lineal model can also retrieve ETC and the ozone absorption coefficient from the comparison with the reference (ETCs,O3ABSs,K,S). In this case the correction is zero for m=1. Indeed we perform these calculations and use the comparison witht the ETCo, and as a check of the calibration. The effect on the stray light parameters are small.

Using the ETC (and the ozone absorption coefficient) from the one point calibration, different from the calculated of the non-linear model produces an generally small offset. In most of the cases the difference is very small,on the worst cases produce an offset 0.3% (Figure 2) on low OSC due to the difference between the ETC/absorption coefficient determined by the operational method and that derived from the fit.

Comment: page 7 Eq. 10: since k is negative, if we want the ozone to increase at the next interaction, there must be a "-" sign, unless alpha is defined negative (Eq. 3). Please, clarify;

Answer: The sign was incorrect ,and corrected now.

Comment: page 7 line 5: "2000 DU" -> to what air mass does it correspond, in El Arenosillo? Is the calculation of the air mass reliable at this SZA? Notice that the stray light correction presented in the paper depends on the product X*mu: if X is low during the determination of the correction, mu must be large (but still accurate!) to cover a sufficiently wide range of OSC, since, at high latitudes, the same OSC can be reached at lower mu's;

Answer: In most of the Brewer spectrometers there is a physical limitation of the observation to 80º, which corresponds to an air-mass around 5 and OSC of 1600 DU during the campaign. The measurement schedule has been adapted to have as much as possible measurements at short angles.

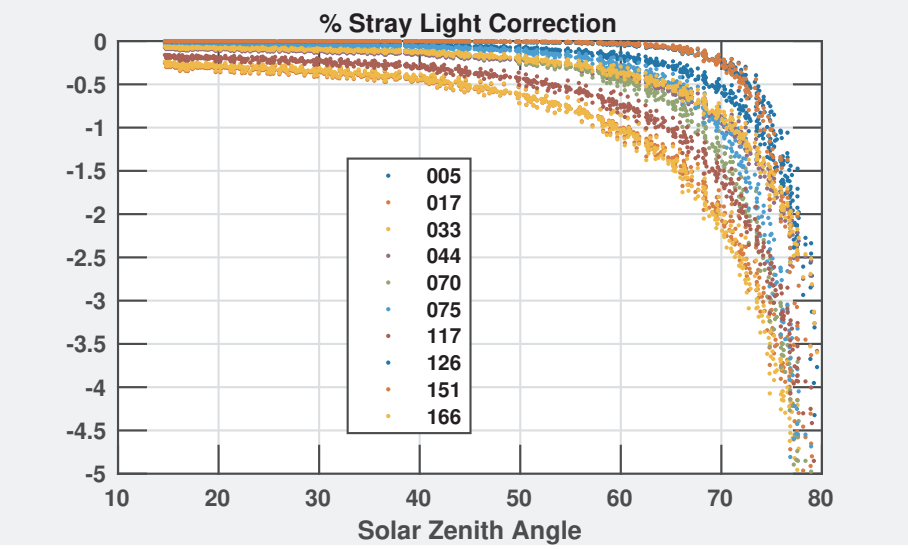

Figure 2. Stray Light correction when the ETC is also derived of the fit, there is an ofset at low OSC due the different ETC but the effect on the stray light constants are small.

Comment: page 7 "3.1 Reference Calibration" -> or "Agreement between reference instruments"?

Answer: Changed and added reference.

Comment: page 8 l. 1-6: the comparison of the reference instruments is an interesting topic, and the RBCC campaigns a very unique chance to investigate it. However, the authors should discuss it with more details. E.g., how are the ECCC Toronto Triad and #158 calibrated? The procedure is explained for the IZO triad, but not for other reference instruments;

Answer: This specific topic is discussed in the work of León-Luis et al. (2018) where different triads are compared, we add in the text a reference for this work

Comment: page 8 l. 5-6: why two different thresholds (900 DU and 600 DU)?

Answer: The #017 is a special case of an instrument with strong stray light, the statistics are calculated up to 900DU, but the underestimation of the ozone is noticeable after 600 DU.

Comment: page 8 l.8 to page 9 l. 7: this basically theoretical part should be described in the "Methods" section, not in the "Results";

Answer: moved.

Comment: page 9 l. 13: Brewer #165 does not exist in the list of the instruments;

Answer: corrected.

Comment: page 10 l. 3: "Brewer #151 can not be considered an operational instrument" -> what does this sentence mean?

Answer: Not all participating instruments are operative, as we include in the text , the non-operational instruments are the instruments that can't provide reliable data and have to be fixed during the campaign. Like the case of #151 that was fixed during the campaign.

Comment: page 10 l. 4: "ago" -> "before";

Answer: The word was changed in the text.

Comment: page 10 l. 12 and 14: "more than 5 units during this period" repeated twice;

Answer: Yes, we have deleted this duplicate phrase.

Comment: page 11 Fig. 9: ordering the x-axis by serial number would improve readability of the chart;

Answer: Done as suggested.

Comment: page 12 Fig. 10: both the bias and the spread for Brewers 158 and 228 is large (in the boxplot). Is there any connection between bias and spread (e.g., instability of the lamps...)?

Answer: There is no relation between bias, which gives us an idea of the instrument change between calibration, and the spread during the campaign periods.

Comment: page 12 l. 1-5: move these lines to a "Methods" section;

Answer: These sentences were moved following the suggestion of the referee.

Comment: page 13 l. 4: if the discussed results were representative of the overall Brewer network, this would mean that about 25% of the instruments are "out of spec" after two years. This is a strong statement: could the author elaborate on this?

Answer: This is what the data suggest, and this is the reason why a two-year calibration is advised. - The overall +/- 1% agreement of the Brewer network as is assumed by the community has been established by the triads of well maintained instrument, but is not done using network instruments. The result is 25 % of the instruments are outside 1% but none of the instruments were outside 1.5%.

Comment: page 13 l. 7: "celebrated" -> "taking place"?

Answer: corrected

Comment: page 14: can the authors draw more definitive and general conclusions from the study?

Answer: We have added the conclusions of the study adding the stray light discussion.

Comment: page 15 Fig. 13: could the variations over time be explained by the variability of the participating instruments? This should be mentioned in the text. Caption: what does "no operating" mean?

Answer:

Yes In effect the variability can be due that the participating instruments are not always the same and can reflect the variability some Brewers only participated in 1-2 campaigns their results may affect overall agreement spread.

Notes

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