

Stability of the RBCC-E Triad during the period 2005 – 2016

Sergio F. León-Luis^{1,2}, Alberto Redondas^{1,2}, Virgilio Carreño^{1,2}, Javier López-Solano^{2,3},
Alberto Berjón^{2,3}, Bentorey Hernández-Cruz^{1,3}, and Daniel Santana-Díaz^{2,3}

¹Izaña Atmospheric Research Center, Agencia Estatal de Meteorología, Tenerife, Spain

²Regional Brewer Calibration Center for Europe, Izaña Atmospheric Research Center, Tenerife, Spain

³Departamento de Ingeniería Industrial, Universidad de La Laguna, Tenerife, Spain

Correspondence to: Alberto Redondas (aredondasm@aemet.es)

Answer to Tom McElroy

Thank you for your review. According to your suggestions and those of other referees, we submit a revised version of our manuscript entitled “Stability of the Regional Calibration Center for Europe Triad during the period 2005-2016” by Sergio Fabián León-Luis, Alberto Redondas, Virgilio Carreño, Javier López-Solano, Alberto Berjón, Bentorey Hernández-Cruz and
5 Daniel Santana-Díaz. All issues brought up by the reviewers have been addressed.

Figures

In all the figures a colour legend has been introduced. For this reason, these are slightly different to the previous version of the article submitted.

Section 2 "Theoretical approach"

10 All referees have indicated that the section 2 is confused. Therefore, it has been rewritten taking into account all suggestions indicated by the referees. Also, a new figure has been introduced.

Grammatical errors

The authors appreciate the grammatical corrections indicated which have been introduced in the text. A co-worker, who is a native English speaker, has helped us with the use of language. In the new version, some paragraphs and sentences were
15 modified to get a more fluid text.

We have were used both “behavior” and “behaviour”. In order to have a more consistent article we have replaced the word “behavior” by “behaviour” in the text.

-Similarly, "tractability" by "traceability". -Similarly, "centre" by "center".

Page 1 Line 1 “Total ozone column measurements can be made using ...” **The text was modified**

- Page 1 Line 2 “In 2003, ...” **The text was modified**
- Page 1 Line 3 “... and since 2011 it has transferred ...” **The text was modified**
- Page 1 Line 4 “... calibration, mainly to other European Brewers, using Brewer #185 ...” **The text was modified**
- Page 1 Line 5 “... annually, alternating ...” **The text was modified**
- 5 Page 1 Line 6 “... focused on reporting on the stability...” **The text was modified**
- Page 1 Line 10 “... to the data from the Izana triad allows the comparison of the ...” **The text was modified**
- Page 1 Line 13 “... standard deviation of the mean equal ...” **The text was modified**
- “In opposite, using...” Suggest “... Alternatively, using the procedure used to analyze the data from the Arosa Triad ...” **The text was modified**
- 10 Page 1 Line 14 “... presents a relative standard deviation of about ...” **The text was modified**
- Page 1 Line 15 “... method used for the data from the World Triad Reference ...” **The text was modified**
- Page 1 Line 16 “... gives monthly values of 0.3% ...” **The text was modified**
- Page 1 Line 17 “... datasets were analyzed ...” “... second only included...” **The text was modified**
- Page 1 Line 18 “Furthermore, this paper also describes the Langley method used to determine the Extra-Terrestrial Constants
- 15 (ETC) for the RBCC-E Triad, the necessary first step toward accurate ozone measurement.” **The text was modified**
- Page 2 Line 2 “Until a few decades ago, it was thought that the ozone concentration was constant in the stratosphere. However, after the discovery of the hole in the ozone layer in the mid-1980s, this idea was discarded (Farman et al., 1985)”. This is a problematic statement. Ozone is variable, it has annual cycles - and longer-term cycles - and a non-uniform global distribution. It is not clear how to accurately express the simple thought the authors are searching for. Perhaps something like
- 20 “Historical measurement s - pre 1980 - indicated that the morphology of ozone was not changing significantly with time. However, the Antarctic measurements of Farman et al., published in 1985, changed that view...” **The text was modified**
- Page 2 Line 4 “Concerns related to the negative effects that UV radiation can have on terrestrial life ...” **The text was modified**
- Page 2 Line 6 “... agents that led to this decrease ...” **The text was modified**
- 25 Page 2 Line 8 “.. total ozone column abundance ...” **The text was modified**
- Page 2 Line 9 “... Fioletov et al., 2005.” And reference: Fioletov, V. E.; J. B. Kerr, C.T. McElroy, D.I. Wardle, V. Savastiouk, and T.S. Grajnar, The Brewer reference triad, Geophys. Res. Lett., 32, 10.1029, 2005. **The reference was modified**
- Page 2 Line 12 Fioletov, V. E.: Comparison of Brewer ultraviolet irradiance measurements with total ozone mapping spectrometer satellite retrievals, Optical Engineering, 41, 3051, <https://doi.org/10.1117/1.1516818>, <http://opticalengineering.spiedigitallibrary.org>.
- 30 This is not a good reference for Brewer aerosol measurements. It is, however, appropriate for UV Irradiance measurements. Suggest that the references be segregated and repeated if necessary to make this clear. **The reference was relocated**
- Page 2 Line 14 “... wavelength separation...” Spelling: “... wavelength separation...” **The text was modified**
- Page 2 Line 16 “Although the prototype Brewer was developed in the early 1970s ...” “... has had on-going technical improvements to improve its accuracy...” **The text was modified**
- 35 Page 2 Line 20 Delete “concentration” **The text was modified**

- Page 2 Line 21 Capitalize “Brewers present this ...” **The text was modified**
- Page 2 Line 24 “... consisting of. ...” **The text was modified**
- Page 2 Line 26 “... (Fioletov et al., 2005) ...” **The text was modified**
- Page 2 Line 27 “... parallel with the World Triad Reference ...” **The text was modified**
- 5 Page 2 Line 30 “... are calibrated by comparison with the travelling ...” **The text was modified**
- Page 3 Line 9 “... Observatory is located on the island of Tenerife ...” **The text was modified**
- Page 3 Line 15 “... campaigns through the travelling... “ **The text was modified**
- Page 3 Line 16 “These values were calculated using measurements in a range where Brewer #017 measurements are not strongly affected by stray light ...” **The text was modified**
- 10 Page 3 Line 23 “... each Brewer at its local station ...” **The text was modified**
- Page 3 Line 32 “... which will allow the calculation of the TOC in near real time ...” **The text was modified**
- Page 4 Line 1 “Currently, approximately 40 Brewers ...” **The text was modified**
- Page 4 Line 9 “Also in this section the results of a study on the behavior of the RBCC-E Triad as a function of SZA range at which the measurements were performed.” **The text was modified**
- 15 Page 4 Line 13 “... four spectral bands which ...” **The text was modified**
- Page 4 Line 14 “... with local maximum and minimum ozone absorption cross-sections.” **The text was modified**
- Page 4 Line 16 “... the contribution of Rayleigh ...” **The text was modified**
- Page 4 Line 17 “... more than one band is ...” This is not the main reason. The ratio technique eliminates the dependence of the absorption function on absolute intensity signal and the right wavelength weighting stops aerosol from interfering.” 17 and
- 20 18 need a little more work. A fifth wavelength is included to measure SO2 **This section was modified**
- Page 4 Line 19 Suggest: “The Beer-Lambert law ...” **The text was modified**
- Page 4 Line 22 “where J is the gas concentration ...” 1. J is used in the literature for optical depth, 2. X is normally used for ozone amount, not in concentration (molecules/cm³) but in cm (or other length units) to go along with " in cm⁻¹. The air mass for ozone is usually written as : to distinguish it from the air mass for Rayleigh scattering, m. **The section 2 was modified and**
- 25 **your suggestions were added**
- Page 4 Line 29 The equations in the PDF did not fare well. **The section 2 was modified and your suggestions were added**
- Page 4 Line 15 Measurements on slits 2 to 6 are used for SO2 and ozone. **The section 2 was modified and your suggestions were added**
- Page 4 Line 24 See earlier notes on variable names. **The section 2 was modified and your suggestions were added**
- 30 On behalf of the co-authors, thank you very much for your suggestions.
Alberto Redondas Marrero.
Regional Brewer calibration Center.
Izaña Atmospheric Research Center

AEMET- Meteorological State Agency, Spain.

Stability of the RBCC-E Triad during the period 2005 – 2016

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Correspondence to: Alberto Redondas (aredondasm@aemet.es)

Answer to Referee #1

Thank you for your review. According to your suggestions and those of other referees, we submit a revised version of our manuscript entitled “Stability of the Regional Calibration Center for Europe Triad during the period 2005-2016” by Sergio Fabián León-Luis, Alberto Redondas, Virgilio Carreño, Javier López-Solano, Alberto Berjón, Bentorey Hernández-Cruz and

5 Daniel Santana-Díaz. All issues brought up by the reviewers have been addressed.

Figures

In all the figures a colour legend has been introduced. For this reason, these are slightly different to the previous version of the article submitted.

Section 2 "Theoretical approach"

10 All referee have indicated that the section 2, where an approach to ozone retrieval (DS routine, slits) and Langley-technique are introduced, is confused. Therefore, it has been rewritten taking into account all the suggestions. Also, a new figure has been introduced.

Grammatical errors

15 The authors appreciate the grammatical corrections indicated which have been introduced in the text. A co-worker, who is a native English speaker, has helped us with the use of language. In the new version, some paragraphs and sentences were modified to get a more fluid text .

We have used both “behavior” and “behaviour”. In order to have a more consistent article we have replaced the word “behavior” by “behaviour” in the text.

-Similarly, "centre" by "center". - P1, 19: add ...from “two different” methods previously... **done**

- P1, 110: it should already be mentioned here that the World Reference is the Toronto triad. **done**
- P2, 15: “on” instead “in”. **done**
- P2, 16: “cause” instead “produce”. **done**
- P2, 19 and in references P20, 14: “Varotsos” instead “varotsos”. **done**
- 5 - P2, 19: “.which are considered to be reference instruments. . .” Is not correct. The triads or Brewer #17 are references (or standards). May be “basic instruments in the global network” or similar is better. **done**
- P2, 110: “Brewer spectrophotometers are widely used” since when? **This sentence was rewritten.**
- P2, 112/13: “ozone concentration” is not correct, “TOC” is better. **done**
- P2, 116: Why is “although” used? Better “After the development of the first Brewer in the early 1980s it has had continu-
- 10 ous...”**This sentence was rewritten.**
- P2, 121: 70 degrees as limit for single Brewer observations seems small, as it correspondsto μ -values of smaller than 3. Even single Brewers can measure reliable TOC up to 75 degrees (μ -values around 3.5) under normally clear air conditions. Perhaps it would be good, to describe the different stray light issues: internal straylight-problem, which is larger for single Brewers, and external stray light (diffuse sky light around the sun), which is similar for all Brewers, but larger for Dobsons.
- 15 The effect is the same in both cases: drop of TOC, when the SZA (μ) increases and gets large depending on instrument and turbidity. **This sentence was rewritten. We think that is better to say that “.. decrease in the TOC measurement at large ozone slant column”**
- P2, 124: The calibration of the Brewer “in the first years” is traceable.
- P2, 131: by the “manufacturer” Kipp & Zonen.**done**
- 20 - P3, 15: Add “triad” behind RBCC-E.**done**
- P3, 19: Replace “in the island” with “on the island”.**done**
- P3, 116: “These values have been”.**done**
- P3, 120: “more than 150 Brewers have been calibrated”. **done**
- P4, 113 ff, p5: see under general comments (SO2!). The reference Dobson 1957 is out of place here and I think anywhere
- 25 in the text. So omit it in the references. The presentation of the corresponding absorption spectra (O3 and SO2) might be helpful.**This section was rewritten**
- P5, 115: DS observations are done in 6, not in 7 slits: 4 ozone, 1 SO2 and 1 dark, the seventh slit is for HG-test. **This section was rewritten**
- P4/5/6: Use of m instead of μ is not correct. **done**
- 30 - P6, 17: “clear” instead of “clean”-sky.**done**
- P6, 111: Why does the use of 1/m (should also be μ) allow obtaining two ETCs? I thought it is the splitting in fore- and afternoon observations.This section was rewritten
- P6, 1 32-35: no complete sentence. **The sentence was corrected**

- P8, 16: Giving a number for the low standard deviation would be helpful.**done** - P8, 19: “Langley technique can be used” instead of “Langley-technique is used”.**done**
- P8, 118: correct “a an”.**done**
- Chapter 4: - P10, 4: “mean of daily differences” or “daily mean of the difference”? **This sentence was rewritten**
- 5 - P10,15: Replace “analyzed” with “analyze”.**done**
- P10, 1 10; replace “inconvenient” with “inconvenience”.**done**
- P10, 111 and P11, 111: isn’t Izana a subtropical station? **This sentences were rewritten**
- P10, 115: “another” instead of “other”.**done**
- P11, figure 4: The “A” from Eq. 10 is missed. Shouldn’t it be placed behind “ozone reference value”? The colors are not
- 10 assigned to the Brewers.**done**
- P11, last section, P12, first section: Sorry, but I have problems to understand what is meant. The beginning of two consecutive sentence with “Therefore” does not sound good. It is described that the polynomial method show similar results regardless of the data set and the order of the fit. Why is then the daily mean method more appropriate? **This paragraph was rewritten**
- P15, Figure 6: The order of the panels does not coincide with the order of the mentioned methods in the figure caption.
- 15 **done**
- P15, 110: correct “surprising”. **done**

Answers at your specific questions:

Comment: The description of the triad system is not profound and clear. Where is the second triad located? In Toronto too? How are this triad and in addition the travelling Brewer No. 17 calibrated? It is good to know how stable the RBCC-E triad is, but what about its accuracy? The agreement between this triad and the World Reference Triad is confirmed by the 0.5% - agreement with #017. But how good is the agreement of #017 with World Reference Triad? When was its last absolute Langley calibration performed?

Answer: In the introduction we made reference to that both (World Reference and the Second Triads) work in parallel in Toronto. In order to clarify this questions, a new reference has been included in the text. An oral presentation where is shown some graphic about the agreement between both triads, and also some photos of the Brewers have been added too. In addition, V. Savastiouk, who operated the Brewer #017, have indicated that “Brewer #017 is regularly compared with the World triad Reference in Toronto it is also absolutely and independently calibrated at the Mauna Loa Observatory in Hawaii, US. Some history of such calibrations can be found at <http://www.io3.ca/Calibrations/Brewer/017>”.(see interactive discussion of our article.)

Comment: P2, 121: 70 degrees as limit for single Brewer observations seems small, as it corresponds to μ -values of smaller than 3. Even single Brewers can measure reliable TOC up to 75 degrees (μ -values around 3.5) under normally clear air conditions. Perhaps it would be good, to describe the different stray light issues: internal straylight-

problem, which is larger for single Brewers, and external stray light (diffuse sky light around the sun), which is similar for all Brewers, but larger for Dobsons. The effect is the same in both cases: drop of TOC, when the SZA (μ) increases and gets large depending on instrument and turbidity.

Answer: Yes, perhaps this value is a very extreme lower limit. This sentence was rewritten. We think that is better to say that "...decrease in the TOC measurement at large ozone slant column".

Comment: - P8, l6: Giving a number for the low standard deviation would be helpful.

Answer: Following the suggestion of other referee this sentence was rewritten "despite this annual behaviour, the ozone presents a lower daily variability as indicated our measurements".

Comment: - P8, l17: The criterion lower than 0.6 under item 4 is not clear: standard deviation of 0.6 DU? Seems to be a very low standard deviation for a day with small ozone variation.

Answer: Yes, this condition was introduced in the article by mistake. It was written in a previous version (internal draft) but it was not deleted from the uploaded article.

Comment:

- P8, l20: what does "(condition 3 above)" mean in the context with simultaneous? Does it mean, that in addition to condition 3, these selected measurements should be simultaneous?.

Answer: rephrased as additional condition.

Comment: - P10, l8/9: what does slight mechanical miscalculation mean?

Answer: Sorry, maybe this sentence is not the best translation into English. The text has been modified as follows:

Old version: "... it should be noted that the presence of slight mechanical miscalculations in the instrument. ..."

New Version: "... it should be noted that the presence of small drifts by its continued operation of the instrument. ..."

Comment: - P11, figure 4: The "A" from Eq. 10 is missed. Shouldn't it be placed behind "ozone reference value"? The colors are not assigned to the Brewers

Answer: A new figure with a colour legend has been upload. The "A" was included in the figure caption.

Comment: - P11, last section, P12, first section: Sorry, but I have problems to understand what is meant. The beginning of two consecutive sentence with "Therefore" does not sound good. It is described that the polynomial method show similar results regardless of the data set and the order of the fit. Why is then the daily mean method more appropriate?

Answer: This paragraph was rewritten because, as you indicate, it is confusing. The "therefore" were replaced by other connectors.

Comment: P13, Fig.5: Again the colors are not assigned to the Brewers

Answer: A new figure with a colour legend have been upload.

Comment: - P12 – P15: What about Brewer #183? Tables 3 and 4 (Data set 1) and Figures 5 and 6 show a larger ratio in 2007, which cannot be seen in the graph of Arosa method. Only the table 4 gives a negative shift median and larger percentile numbers for Brewer #183 data set 1. Is the reason known? The same is valid for the larger scattering of all RBCC-E Brewers around 2010.

Answer: Thank you! If the data in this graph was wrong. We have reviewed our calculations and now the results are more coherent with the previous graphs.

Comment:

2010: issues on 183, during that time only 157 and 185 were used for reference.

Answer: In 2010, the brewer #183 had some problem with their micrometer during some months. As in the previous questions, the fit can hide this problem. We have introduced this reference where some plot about this are shown. *Roozendaal, M. V., Köhler, U., Pappalardo, G., Kyrö, E., Redondas, A., Wittrock, F., Amodeo, A. and Pinardi, G.: CEOS Intercalibration of Ground-Based Spectrometers and Lidars: Final report. <http://repositorio.aemet.es/handle/20.500.11765/8886> (Accessed 5 April 2018), 2013.*

Comment: - P14, I9 (and in abstract too): Where do the numbers for the RBCC-E triad come from? I cannot find them in the tables.

Answer: The values are the serial numbers of the brewer

Comment: - P14, I10: how is 40% lower dispersion determined?

Answer: This value was calculated as the ratio between both relative deviation $(0.47-0.27)/0.47=0.42$

Comment: - P14, I12: Is the 0.3% value for RBCC-E also calculated in the 2004 – 2012 period of the Stübi-paper

Answer: We're sorry, but we cannot calculate that data. As we indicated in the introduction, brewers # 183 and # 185 were installed in 2005, hence there is not available data in the year 2004. In the article, the value reported corresponds with the period 2005-2016. In addition, it can be confusing to give values that do not correspond with the years defined in the datasets.

Comment: - P15, Figure 6: The order of the panels does not coincide with the order of the mentioned methods in the figure caption.

Answer: We have rewritten the Figure caption in the correct order.

Comment: - P15/16: The explanation for the larger scattering at high sun is good and comprehensible. But the obvious difference of low sun in the forenoon and in the afternoon is neither mentioned nor explained. Is there also an

explanation, why Brewer #183 shows larger scattering in the relative difference than the other two Brewers?

Answer: Honestly, we think that the Brewer \#183 presents this large scattering because it was damaged in 2007, during the “Delta” storm and during 2008 it had an irregular behaviour and this data was used in this analysis. In addition, this difference is minimal or negligible and does not affect the calibration of the Triad. Moreover, this result is consistent with those presented in tables 3 and 4 where it is found that brewer 183 has the poorer values. It seems logical to think that if in average values (daily or monthly) this brewer is the worst, also depending on the SZA this difference is reflected.

Comment: - P16/17: The values in table 5 are not clear. Where do they come from? Fioletov daily of 0.47% is mentioned on P16, 18. In the same context the RBCC-E triad number is given as 0.41% in the Conclusion, but as 0.37 in the table, which is confusing.

Answer:

In order to clarify this question, a new table was generated.

On behalf of the co-authors, thank you very much for your suggestions.

Alberto Redondas Marrero.

Regional Brewer calibration Center.

Izaña Atmospheric Research Center

AEMET- Meteorological State Agency, Spain.

Stability of the RBCC-E Triad during the period 2005 – 2016

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Answer to V. Savastiouk

Thank you for your review. According to your suggestions and those of other referees, we submit a revised version of our manuscript entitled “Stability of the Regional Calibration Center for Europe Triad during the period 2005-2016” by Sergio Fabián León-Luis, Alberto Redondas, Virgilio Carreño, Javier López-Solano, Alberto Berjón, Bentorey Hernández-Cruz and
5 Daniel Santana-Díaz. All issues brought up by the reviewers have been addressed.

Figures

In all the figures a colour legend has been introduced. For this reason, these are slightly different to the previous version of the article submitted.

General corrections

- 10 The authors appreciate the grammatical corrections indicated which have been introduced in the text. A co-worker, who is a native English speaker, has helped us with the use of language. In the new version, some paragraphs and sentences were modified to get a more fluid text.
- We have used both “behavior” and “behaviour” words. In order to have a more consistent article we have replaced “behavior” by “behaviour” in the text. Similarly, “centre” by “center”.
- 15 - In addition, the expression “ozone concentration” was replaced by “Total ozone column (TOC)”.
- The word “lines” was replaced by “bands” to indicate the wavelengths where the brewer measured.

References

New references have been included in the text where you and the other referees have indicated. However, we think that introducing a reference in the abstract as you suggest in your report is not habitual.

Abstract

P1- L9. This sentence was rewritten taking into account your suggestions. Also, the standard deviation symbol σ has been introduced together with the values reported.

Introduction

5 **P2- L5.** The sentence where we suggest that "... several countries agreed to reduce the agents that produces the decrease of ..." was modified and, the number of countries agreed (197) was included.

P2- L12. The expression "Brewer is a spectrophotometer" was replaced by "Brewer ozone spectrophotometer". Also, this paragraph were rewritten.

10 **P2- L21.** 70 degrees as limit for single Brewer observations seems small, as it corresponds to -values of smaller than 3. We think that is better to say that " The single Brewer presents this problem for large ozone slant column (OSC)".

P2- L32. We think that is correct to duplicate the sentence "Since November 2003 and within the World Meteorological Organization (WMO) and the Global Atmosphere Watch " because in this paragraph it is used to talk about the history of the RBCC-E and, obviously, you must say: when was created it?, when the brewers were installed ?, which is our travelling Brewer?, etc.

15 **P3- L11** We say that Izaña has "clean air and clear sky conditions around all the year and offers excellent conditions to perform the Langley-technique". As you indicate we have not taken into account the dust intrusions. Therefore, the sentence was rewritten: "This ensures clean air and clear sky conditions around all the year and offers excellent conditions to perform the Langley technique, except for some days where the Saharan dust intrusions make difficult to measure the direct solar radiation".

20 **P3- L12.** The sentence "Moreover, comparisons with the World Triad reference are carried out regularly..." was rewritten as " Moreover, the traceability between the RBCC-E Triad and the World Triad Reference is checked during the calibration campaigns through the travelling references #185 and #017."

P3- L19. The expression "annually alternating" was replaced by "annually, alternating"

P3- L23. The sentence "... on the measurements performed by each Brewer in its local station" was rewritten.

25 **P4- L4.** The sentence "... the precision between the measurements" was rewritten as "how similar are the measurements made by the Brewers "

Theoretical Approach

All referees have indicated that the section 2 is confused. Therefore, it has been rewritten taking into account all the suggestions about the approach to ozone retrieval (DS routine, slits) and Langley-technique. Also, a new figure has been introduced and the
30 equations were modified, adding the symbol μ . In addition, a clearer explanation about the procedure of the Langley technique was written. In it, we relate the Langley with the inverse of μ , and also explain how it affects the aerosols in this technique.

Now, we hope that the order of the section will be better.

Ozone and dataset selected

P8- L6. The sentence "Despite this annual behaviour, the ozone is stable during the day, with a low standard deviation for the recorded data." was rewritten as "Despite this annual behaviour, the ozone presents a lower daily variability as indicated by our
5 measurements".

P8- L17. The criterion lower than 0.6 under item 4 is not clear: standard deviation of 0.6 DU? Seems to be a very low standard deviation for a day with small ozone variation.

Yes, this condition was introduced in the article by mistake. It was written in a previous version (internal draft) but it was not deleted from the uploaded article.

10 Results and discussion

P10- L8/9. what does slight mechanical miscalculation mean?

Sorry, maybe this sentence is not the best translation into English. The text has been modified as follows:

Old version: "... it should be noted that the presence of slight mechanical miscalculations in the instrument. ..."

New Version: "... it should be noted that the presence of small drifts by its continued operation of the instrument. ..."

P12 -L12. The equation "mean value" was correct.

P10- L15. The expression "our experience suggest" was rewritten as "our experience suggests".

Conclusions

Some sentences were rewritten. The standard deviation symbol σ has been introduced to identify the values reported and a new table where all values are summarised is also included.

Stability of the [..*]Regional Brewer Calibration Center for Europe Triad during the period 2005 – 2016

Sergio Fabián León-Luis^{1,2}, Alberto Redondas^{1,2}, Virgilio Carreño^{1,2}, Javier López-Solano^{1,2,3}, Alberto Berjón^{2,3}, Bentorey Hernández-Cruz^{1,2,3}, and Daniel Santana-Díaz^{2,3}

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Correspondence to: Alberto Redondas (aredondasm@aemet.es)

Abstract. Total ozone column [..²]measurements can be made using Brewer spectrophotometers which are calibrated periodically in intercomparison campaigns with respect to a reference instrument. In 2003, the Regional Brewer Calibration Centre for Europe (RBCC-E) was established at the Izaña Atmospheric Research Centre (Canary Islands, Spain) and [..³]since 2011 it [..⁴]transfers its own calibration, mainly to other European Brewers, using the Brewer #185 as [..⁵]travelling reference.

- 5 This work is focused on [..⁶]reporting on the stability of the measurements of the RBCC-E Triad (Brewers #157, #183 and #185) [..⁷]made at the Izaña Atmospheric Observatory during the period 2005 – 2016. In order to study the long-term [..⁸]precision of the RBCC-E Triad, it must be taken into account that each Brewer performs a large number of measurements every day and, hence, it becomes necessary to calculate a representative value of all of them. This value was calculated from two different methods previously used to study the long-term [..⁹]behaviour of the World Reference [..¹⁰]Triad (so-called
- 10 Toronto Triad) and Arosa Triad. Applying their procedures [..¹¹]to the data from the RBCC-E Triad allows the comparison of the three instruments. In [..¹²]daily averages, applying the procedure used for the World Triad Reference, the RBCC-E Triad presents a relative standard deviation equal to $\sigma=0.41\%$ which is calculated as the mean of the individual values for each Brewer [..¹³]($\sigma_{157} = 0.362\%$, $\sigma_{183} = 0.453\%$ and $\sigma_{185} = 0.428\%$). Alternatively, using the procedure used to analyze

*removed: RBCC-E

²removed: can be measured

³removed: from

⁴removed: has transferred

⁵removed: reference instrument. The RBCC-E organizes regular inter-comparisons which are held annually alternating between Arosa (Switzerland) and El Arenosillo (Spain).

⁶removed: showing

⁷removed: in the

⁸removed: stability

⁹removed: stability

¹⁰removed: and Arosa Triads

¹¹removed: in our triad allows us to compare the

¹²removed: this way, the difference between the values calculated

¹³removed: and the triad mean was analyzed. This also allows us to quantify the precision of each Brewer. The results obtained show that these differences are lower than 2 Dobson units in approx 90% of the days evaluated. The daily and monthly difference between each Brewer and the Triad were calculated. We analyze

the Arosa Triad, the RBCC-E presents a relative standard deviation of about $\sigma = 0.5\%$. In monthly averages, the method used for the data from the World Triad Reference give a relative standard deviation mean equal to $\sigma=0.3\%$ ($\sigma_{157} = 0.33\%$, $\sigma_{183} = 0.34\%$ and $\sigma_{185} = 0.23\%$). Whereas, the procedure of the Arosa Triad gives monthly values $\sigma=0.5\%$. In this work, two ozone datasets are analyzed: the first [..¹⁴]include all the ozone measurements available while the second only includes the simultaneous measurements of all three instruments. Furthermore, [..¹⁵]this paper also describes the Langley method used [..¹⁶]to determine the Extra-Terrestrial Constant (ETC) [..¹⁷]for the RBCC-E Triad, the necessary first step [..¹⁸]toward accurate ozone measurement. Finally, the short-term, or intraday, stability is also studied to identify the effect of the solar zenith angle on the accuracy of the RBCC-E Triad.

1 Introduction[..¹⁹]

The ozone layer is a region of the Earth's stratosphere that absorbs most of the Sun's Ultraviolet (UV) radiation (Anwar et al., 2016). Historical measurements -pre 1980- indicated that the morphology of ozone was not changing significantly with time. [..²⁰]However, [..²¹]the Antarctic measurements of Farman et al. (1985) changed that view. Concerns related to the negative effects that UV can have on terrestrial life led to the signing of the Montreal Protocol in 1987, where [..²²]197 countries agreed to reduce the agents that [..²³]led to this decrease (Sarma and Andersen, 2011). From this date, the monitoring and control of the [..²⁴]total ozone column abundance has been a priority of the World Meteorological Organization (WMO). This task requires instruments that can measure the total ozone column concentration with [..²⁵]an accuracy of $\sim 1\text{-}3\%$ such as the Dobson and Brewer which are considered as [..²⁶]ideal instruments for monitoring the ozone abundance (Basher, 1985; Varotsos and Cracknell, 1994; Fioletov et al., 2005; Scarnato et al., 2009).

Brewer [..²⁷]ozone spectrophotometers are used to measure the total ozone column (TOC), ultraviolet irradiance (Fioletov, 2002) and, more recently, the aerosol optical depth in the ultraviolet range [..²⁸](Carvalho and Henriques, 2000; Gröbner et al., 2001; López-Solano et al., 2017). This instrument is mounted on an azimuth tracker that determines the [..²⁹]TOC

¹⁴removed: included

¹⁵removed: in this paper we also describe

¹⁶removed: in the RBCC-E Triad to calculate the Extra-terrestrial constant

¹⁷removed: , which is

¹⁸removed: to ozone retrieval

¹⁹removed: .

²⁰removed: Until a few decades ago, it was thought that the ozone concentration was constant in the stratosphere.

²¹removed: after the discovery of the hole in the ozone layer in the mid-1980s, this idea was discarded (Farman et al., 1985). The negative impact that ultraviolet radiation has in

²²removed: several

²³removed: produce

²⁴removed: ozone layer

²⁵removed: great accuracy

²⁶removed: reference instruments

²⁷removed: spectrophotometers are widely

²⁸removed: (Carvalho and Henriques, 2000; Gröbner et al., 2001; Fioletov, 2002; López-Solano et al., 2017). The Brewer is a spectrophotometer mounted on a sun

²⁹removed: ozone concentration

from a direct measurement of the solar radiation. ^[..³⁰]The grating system separates the solar radiation and a slit mask mechanism is used to select the different UV ^[..³¹]bands to be measured ^[..³²]which are associated with maximum and minimum ozone absorption ^[..³³]cross-sections.

- ^[..³⁴]After the development of the first Brewer in the early ^[..³⁵]1970s (Brewer, 1973; Kerr et al., 1985), it has had ^[..³⁶]on-going technical improvements to ^[..³⁷]improve its accuracy. For example, in the photomultiplier, diffraction gratings, ^[..³⁸]operating software used as well as the incorporation of new measurement routines ^[..³⁹](Fioletov et al., 2005, 2011; Karppinen et al., 2015; Fountoulakis et al., 2017). However, possibly the greatest improvement has been the transition from a single to a double monochromator. This eliminates the presence of stray light in the measurements ^[..⁴⁰]which causes a decrease in the ^[..⁴¹]TOC at large solar zenith angles (Karppinen et al., 2015). In practice, the single Brewer presents this problem for large ozone slant column (OSC). Although, depending on the instrument, this effect may be greater or lesser (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2015; Redondas and Rodríguez-Franco, 2016).

- The calibration of the Brewer is traceable ^[..⁴²]from the World Triad Reference, managed by Environment and Climate Change Canada, consisting ^[..⁴³]of Brewers #008, #014 and #015, ^[..⁴⁴]and located at Toronto. These single Brewers are calibrated every few years at the Mauna Loa Observatory (Hawaii) using the ^[..⁴⁵]Langley-technique (Fioletov et al., 2005). A second triad formed by double Brewers #145, #187 and #191 is also ^[..⁴⁶]operated in parallel with the World Triad Reference (Natcheva, 2014; Fioletov and Natcheva, 2014; Zhao et al., 2016). Also, the Swiss Federal Office of Meteorology and ^[..⁴⁷]Climatology (Meteo Swiss) has the Arosa Triad formed by the singles (#040 and #072) and double (#156) Brewers (Stübi et al., 2017). However, the Brewers distributed around the World are calibrated by comparison with the travelling standard reference, Brewer #017, managed by International Ozone Services (IOS) and Brewer #158 manufacturer by Kipp & Zonen.

In addition, since November 2003 and within the World Meteorological Organization (WMO) and the Global Atmosphere Watch (GAW) Programme, the Regional Brewer Calibration Centre (RBCC-E) for RA-VI Region was established at the Izaña

³⁰removed: For this, each Brewer has a holographic grating system to split

³¹removed: spectral lines

³²removed: . These lines

³³removed: bands

³⁴removed: Although the first Brewer was developed

³⁵removed: 1980s

³⁶removed: continuous

³⁷removed: gain accuracy. This includes improvements

³⁸removed: and operating software , and also

³⁹removed: (Fioletov et al., 2011; Karppinen et al., 2015; Fountoulakis et al., 2017)

⁴⁰removed: that

⁴¹removed: real TOC concentration

⁴²removed: to the Triad belonging to

⁴³removed: in

⁴⁴removed: which is considered the World Triad Reference

⁴⁵removed: Langley method (Fioletov et al., 2005). However, the Brewers distributed around the World are calibrated from the travelling standard reference, Brewer #017, managed by International Ozone Services (IOS) and Brewer #158 managed by Kipp & Zonen.

⁴⁶removed: operational (Zhao et al., 2016)

⁴⁷removed: climatology

Atmospheric Observatory (IZO) which is located on the island of Tenerife, managed by the ^[..⁴⁸]State Meteorological Agency of Spain (AEMET). The RBCC-E is the European Brewer Reference and hence can calibrate and transfer its own calibration. Its trajectory started in the year 1997 when the first double Brewer #157 was installed at IZO, running in parallel with a single Brewer #033 for six months. In January 2005, a second double Brewer, the #183, was installed and designated as the travelling reference. The single Brewer #033 was moved to Santa Cruz Meteorological Station (^[..⁴⁹]CMT) in December 1997, leaving the RBCC-E with only two instruments. In July 2005, a third double Brewer #185 was installed. Since that moment, the RBCC-E has been formed by the Brewers #157, #183 and #185. The TOC measured by Regional Primary Reference #157 are sent regularly to different world data servers. The Regional Secondary Reference #183 is used to ^[..⁵⁰]developing and test. Whereas, the Regional Travelling Reference corresponds to Brewer #185.

The Izaña Atmospheric Observatory is located ^[..⁵¹]on the island of Tenerife, on the top of a mountain plateau at 2373 m a.s.l. The observatory is thus located ^[..⁵²]on the region below the descending branch of the Hadley cell, typically above a stable inversion layer, and on an island far away from any significant industrial activities. This ensures clean air and clear sky conditions around all the year and offers excellent conditions to perform the ^[..⁵³]Langley-technique, except for some days where the Saharan dust intrusions inhibit the measurements of the direct solar radiation. Each Brewer can be calibrated “in situ” and independently using the ^[..⁵⁴]Langley-technique, without the need to move them to other locations. Moreover, ^[..⁵⁵]the traceability between the RBCC-E Triad and the World Triad Reference ^[..⁵⁶]is checked during the calibration campaigns through the travelling references #185 and #017. In this comparison, both instruments agree within 0.5%. These values have been calculated using measurements in a range where Brewer #017 measurements are not strongly affected by stray light (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2015; Redondas and Rodríguez-Franco, 2016)

The RBCC-E also organizes inter-comparisons which are held annually, alternating between Arosa (Switzerland) and El Arenosillo (Spain). Since 2011, more than 150 calibrations have been ^[..⁵⁷]conduted (Cuevas et al., 2015). In these campaigns, the RBCC-E facilitates a new calibration for each instrument. Moreover, in order to obtain an ozone value with better accuracy, the RBCC-E advises on the need to ^[..⁵⁸]reprocess the observations performed by each ^[..⁵⁹]brewer at its local station ^[..⁶⁰](Redondas et al., 2018). Aside from regular inter-comparisons, the RBCC-E has carried out other research campaigns supported by the ESA CalVal project. The NORDIC campaigns, with the objective to study the ozone measurements at

⁴⁸removed: Agencia Estatal de Meteorología

⁴⁹removed: SCO

⁵⁰removed: check new routines

⁵¹removed: in

⁵²removed: in

⁵³removed: Langley technique

⁵⁴removed: Langley plot method and

⁵⁵removed: comparisons with the

⁵⁶removed: (Cuevas et al., 2015) are carried out regularly.

⁵⁷removed: performed (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2016)

⁵⁸removed: apply some type of correction (normally, standard lamp correction) on the measurements

⁵⁹removed: Brewer in

⁶⁰removed: before the campaigns

high latitudes, and the Absolute Calibration Campaigns performed at IZO with the participation of Brewer and Dobson reference instruments. The participating Brewers and the travelling reference #185 operate with the same schedule throughout these campaigns. The TOC [..⁶¹] recorded by the travelling reference #185 are used to calibrate the participating Brewers and also to conduct research works [..⁶²](De La Casinière et al., 2005; Redondas et al., 2015; Redondas and Rodriguez-Franco, 5 2016).

Finally, it should be also mentioned that within the framework of COST Action ES1207, “A European Brewer Network” (EUBREWNET), the RBCC-E and AEMET are developing a dataserer for EUBREWNET (<http://rbcce.aemet.es/eubrewnet>) which will allow [..⁶³]the calculation of the TOC in near real time (Rimmer et al., 2017). This completes the objectives of this COST action, whose aim is establishing a coherent network of Brewer monitoring stations in order to harmonise operations and develop approaches, practices and protocols to achieve consistency in quality control, quality assurance and coordinated 10 operations. Currently, [..⁶⁴]approximately 40 Brewers, mainly European, send their data automatically every 20 minutes to EUBREWNET’s dataserer. This dataserer also allows the reprocessing of data for homogenization and automatic quality control.

The present work [..⁶⁵]focused on investigating how similar are the measurements made by the Brewers #157, #183 and #185 [..⁶⁶]each day and how stable does this behaviour remain over time. This allows us to identify [..⁶⁷]periods with lower or higher agreement between the Brewers. The RBCC-E measurements are evaluated from the methods described for the World Reference and Arosa Triads to study its stability. With this idea in mind, this work has been structured as follows: an approach to ozone retrieval and Langley method is presented in Section 2. The [..⁶⁸]ozone values recorded in the period 2005-2016 and datasets used are shown in Section 3. The methods used to calculate the daily ozone value [..⁶⁹]and the results obtained from these values and its discussion are presented in Section 4. Also, this section includes results of a study on the behaviour of the RBCC-E Triad as a function of SZA range at which the measurements were performed. 20 Finally, the conclusions are presented in Section 5.

⁶¹removed: concentrations

⁶²removed: (Redondas et al., 2015; Redondas and Rodriguez-Franco, 2016; De La Casinière et al., 2005)

⁶³removed: to calculate the TOC concentration

⁶⁴removed: around

⁶⁵removed: shows the consistence between the measurements performed at IZO

⁶⁶removed: in the period 2005-2016

⁶⁷removed: the years with high or low long-term stability. Also, the accuracy in function of the solar zenith angle (SZA) is studied to evaluate the intraday dependence (short-term stability). This

⁶⁸removed: selection criteria for the datasets evaluated and the procedure

⁶⁹removed: from these are shown in Section 3. The results

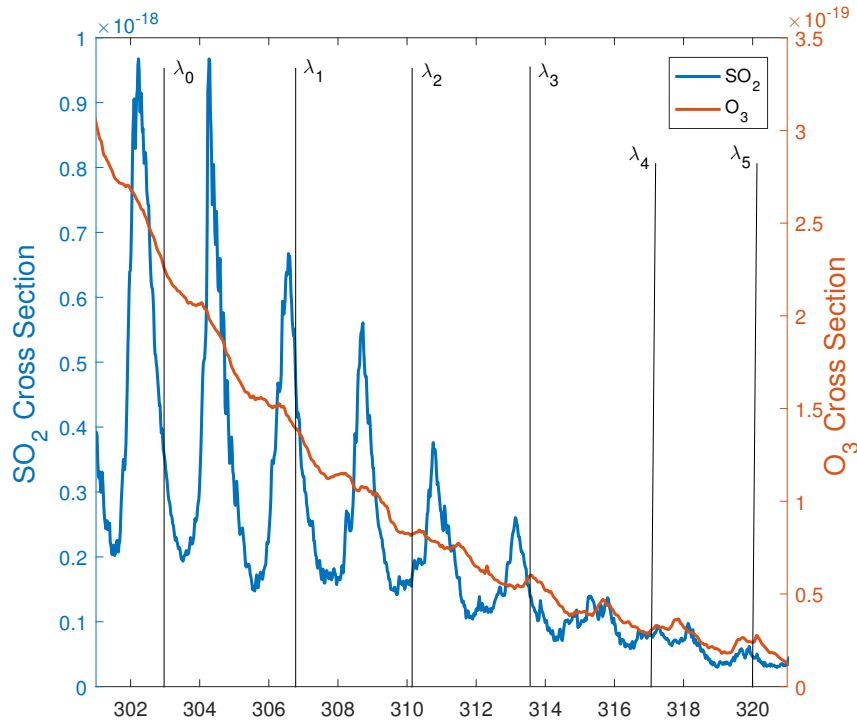


Figure 1. Ozone and sulfur dioxide absorption cross sections. The solar radiation is measured for the intensity bands ($\lambda_{1-5} = 306.4, 310.1, 313.5, 316.8, 320.0nm$). In contrast, the wavelength $\lambda_0 = 303nm$ is used for a check routine.

2 [..⁷⁰]

2 Theoretical Approach

2.1 Ozone retrieval.

[..⁷¹]

- 5 The standard (so-called DS) routine used to determine the TOC from direct sunlight radiation, measuring the signal intensity in five bands ($\lambda_{1-5} = 306.4, 310.1, 313.5, 316.8, 320.0nm$) which are associated with maximum and minimum [..⁷²]
]O₃ and SO₂ absorption cross sections, see Fig. 1. Despite that SO₂ presents a more efficient absorption, its lower presence in the atmosphere (5 D.U.) compared to the ozone (200-500 D.U.) causes that the greater absorption of UV

⁷⁰removed: Theoretical Approach.

⁷¹removed: Each Brewer, in its movement following the Sun, measures the direct solar radiation in four spectral lines

⁷²removed: ozone absorption bands. The line intensity

radiation is due to the latter (Kerr, 2010). The intensity measured F [..⁷³], in raw counts for each wavelength, can be expressed in terms of counts per second, after applying some instrumental corrections [..⁷⁴] (dark counts, dead time [..⁷⁵] and temperature coefficients) and, also, taking into account the contribution of [..⁷⁶] Rayleigh scattering. [..⁷⁷] [..⁷⁸]

5 [..⁷⁹]

[..⁸⁰] Using standard Brewer operational variables, [..⁸¹] the TOC can be obtained as follows,

$$O_3 = [..⁸²] \frac{MS9 - ETC}{\alpha \cdot \mu} \quad (1)$$

where [..⁸³] MS9 [..⁸⁴] (so-called double ratio) is calculated as follows, (Brewer, 1973; Kerr et al., 1981, 1985; Kipp & Zonen, 2008).

$$10 \quad MS9 = 10^4 \sum_i w_i \log F_i = 10^4 (\log F_2 - 0.5 \log F_3 - 2.2 \log F_4 + 1.7 \log F_5) \quad (2)$$

The ozone absorption coefficient, α , [..⁸⁵]

[..⁸⁶]

15 [..⁸⁷]

[..⁸⁸]

[..⁸⁹] is calculated from dispersion test (Redondas et al., 2014a) [..⁹⁰]

$$\alpha = 10^4 \sum_i w_i \log \alpha_i = 10^4 (\log \alpha_2 - 0.5 \log \alpha_3 - 2.2 \log \alpha_4 + 1.7 \log \alpha_5) \quad (3)$$

⁷³ removed:)

⁷⁴ removed: on the raw counts (the so called

⁷⁵ removed: , and temperature corrections) and also

⁷⁶ removed: the

⁷⁷ removed: The main reason why it is necessary to measure more than one line is the overlapping of the O_3 and SO_2 absorption bands. So, it is necessary to measure more than one line to get information about the SO_2 contribution and subtract it.

⁷⁸ removed: The Lambert-Beer's law relates the irradiance at the ground and the top of the atmosphere (F_i and F_i^0 , respectively) for each different wavelengths (denoted by i subindex) as follows:

⁸⁰ removed: where τ is the gas concentration (O_3 , SO_2 , etc) and its absorption coefficient (α_i). The distance traveled by the solar radiation is given by the air mass, m .

⁸¹ removed: Eq. ?? can be solved for the TOC

⁸³ removed: ETC and

⁸⁴ removed: represent the solar radiation measured at the top of the atmosphere and the ground, respectively (Kipp & Zonen, 2008). Both parameters, and also the

⁸⁵ removed: must be understood as values obtained from the weighted linear combination of the intensities F , in logarithmic scale, of the four lines measured:

⁸⁹ removed: The absorption intensity lines α_i are

⁹⁰ removed: .

where α_i represents the band intensity calculated for each wavelength indicated in Fig. 1.

The weights, $w_i = (-1, 0.5, 2.2, -1.7)$ and the wavelengths, $\lambda_i = (310.1, 313.5, 316.8, 320.0nm)$, used have been especially selected to suppress the aerosol and SO₂ effects in the measured signal (Dobson, 1957; Kerr et al., 1981). These w_i and λ fulfill the equations 4 and 5

$$\sum_{i=1}^4 w_i = 0$$

$$\sum_{i=1}^4 w_i \lambda_i \approx 0$$

ensuring that any linear effects with wavelength are suppressed and also allow to minimize any small shift in wavelength and the influence of SO₂ on the ozone retrieval.

$$\sum_{i=1}^4 w_i = 0 \quad (4)$$

$$\sum_{i=1}^4 w_i \lambda_i \approx 0 \quad (5)$$

It is important to note that the factor 10^4 introduced in Eq. 2 is because the Brewer algorithm works in an internal base 10 logarithmic space multiplied by this factor. A more extensive description about dispersion test and the mathematical procedure to calculate the ozone concentration can be found in (Gröbner et al., 1998; Kipp & Zonen, 2008). Finally, the *ETC* (so-called Extra-Terrestrial Constant) must be calculated directly using the Langley-technique or transfers by comparison with a reference instrument.

2.2

2.2 Langley calibration method for the RBCC-E Triad

The Langley-technique is the most popular procedure to estimate

⁹¹removed: λ and
⁹²removed: verify
⁹³removed: .
⁹⁶removed: sulfur dioxide
⁹⁷removed: ??
⁹⁸removed: Also note that the standard (so-called DS) routine used to determine the ozone concentration from direct sunlight radiation measures the signal intensity in seven slits, $i = 0-6$, the second devoted to the dark counts and the remaining ones to the SO₂ and O₃ absorption. This explains the values of the subscripts in Eq. ?. There is a further slit 0, which is used to measure the spectral line of an internal HG lamp as an auto-calibration method. Slit 2 only contains contributions from the SO₂ absorption and is not used in Eq. ?.
⁹⁹removed: Kipp & Zonen (2008); Gröbner et al. (1998).
¹⁰⁰removed: Langley calibration method for the RBCC-E Triad.
¹⁰¹removed: All the parameters in Eq. 1 are known except the solar radiation at the top of the atmosphere *ETC*, which must be estimated. The Langley calibration method
¹⁰²removed: it. This technique is based on Lambert-Beer's law (Eq. ?) written as a linear equation with the total optical air mass m as the independent variable and $\log_e F_i^0$ as the intercept:

[..¹⁰⁴] the extraterrestrial constant *ETC*. In practice, with respect to [..¹⁰⁵] TOC measurements, the [..¹⁰⁶] *ETC* can be calculated directly fitting a linear equation to the *MS9* values respect to air mass [..¹⁰⁷] μ :

$$MS9 = ETC + O_3\alpha\mu \quad (6)$$

or using the Dobson method (Dobson and Normand, 1958; Komhyr et al., 1989):

$$5 \quad [..¹⁰⁸] \frac{MS9 - (ETC + \Delta ETC)}{\mu} = [..¹⁰⁹] O_3\alpha [..¹¹⁰] \quad (7)$$

[..¹¹¹] where ΔETC represents the variation of this parameter respect to a reference value.

Both equations can be used to calculate the *ETC* value to be used in Eq.1 but Eq.7, where the slope is inverse to μ , has the advantage of presenting a better data distribution (Kiedron and Michalsky, 2016). This is because the number of measurements performed at low air mass ($1 \leq \mu \leq 2$) are more than those at large air mass ($\mu \geq 2$). Although all
10 measurements could be used for this calculation, experience suggests that is better to select a subset of measurements. [..¹¹²] The days with a [..¹¹³] high aerosol optical depth concentration, i. e. during Saharan dust intrusions, are removed from this study with the help of data reported by other instruments.

The methodology used is essentially the same as described in (Redondas, 2008; Redondas et al., 2014b). The following criteria, listed in order of application, can be used to get a good agreement between the *ETC* values calculated:

- 15 1. [..¹¹⁴] The regression is performed on the [1.25 , 3.5] airmass range, using the brewer astronomical formulas for the airmass determination.
2. The data recorded during the morning and afternoon are taken separately (2 [..¹¹⁵] Langley per day)
3. [..¹¹⁶] Individual measurements (not the average of 5) are considered with the cloud screen method of 2.5 ozone
20 standard deviation.
4. This daily standard deviation limit (2.5 DU) are used to select the Langley events.

¹⁰⁴removed: This procedure could be applied directly to calculate the solar extraterrestrial constant , in counts per second, for each one of the spectral lines measured, see Eq. ???. However

¹⁰⁵removed: ozone

¹⁰⁶removed: ETC is

¹⁰⁷removed: m ,

¹¹¹removed: Although, in principle,

¹¹²removed: This is because the atmospheric conditions are not stable throughout a day and this variability affects the *ETC* value calculated. Therefore, in practice, the

¹¹³removed: stable atmosphere are selected. Redondas (2008) have shown that the

¹¹⁴removed: Days with a stable ozone concentration (standard deviation lower than

¹¹⁵removed: DU) and a large number of measurements (more than 50) .

¹¹⁶removed: Days with a low aerosol optical depth concentration and clean-sky conditions.

5. MS9 double ratios are corrected for filter non linearity.

[..¹¹⁷] It is important to indicate that despite selecting the better days [..¹¹⁸] when the *ETC* values obtained for different days are compared, [..¹¹⁹] these present a standard deviation around ± 5 . This difference is considered normal and the [..¹²⁰] *ETC* introduced in Eq. 1 corresponds to the [..¹²¹] *ETC* mean. Although the median can be another option to get the

5 *ETC*, the previous criteria guarantee that the difference between both methods are not significantly.

Aside from the interest to determine the [..¹²²] TOC, the *ETC* is considered as a probe to check the correct state of the instrument. [..¹²³] The *ETC* calculated from the Langley method presents a near constant value (std. ± 5), [..¹²⁴] changing only when the instrument [..¹²⁵] recalibrates. This may happen, for example, after replacing a damaged component or due to normal drifts by its continued operation. [..¹²⁶] In both cases, and after a stabilization period, a new [..¹²⁷] *ETC* value can be

10 calculated.

3 [..¹²⁸]

2.1 [..¹²⁹]

[..¹³⁰]

[..¹³¹] As an example, Fig. 2 shows the operative value of the *ETC* for the Brewer # [..¹³²]

15 [..¹³³]

¹¹⁷removed: In this respect, during a large part of the year, the Izaña Atmospheric Observatory presents the ideal atmospheric conditions required to use the Langley calibration method. However, and due to the low latitude of the Canary Islands, the number of measurements performed at low air mass ($1 \leq m \leq 2$) are more than those at large air mass ($m \geq 2$). This produces a non-homogeneous data distribution. For this reason, Redondas (2008) suggest to make a Langley fit in the scale 1/m. This allows obtaining two ETC values per day.

¹¹⁸removed: , when the ETC values obtained in

¹¹⁹removed: its standard deviation is

¹²⁰removed: ETC introduced in the

¹²¹removed: ETC mean.

¹²²removed: ozone concentration, the ETC

¹²³removed: So, in a period with a stable behavior, the ETC

¹²⁴removed: increasing

¹²⁵removed: losses its calibration

¹²⁶removed: On

¹²⁷removed: ETC

¹²⁸removed: RBCC-E Triad stability: Ozone and dataset selection criteria.

¹²⁹removed: Representative value of the Total ozone column.

¹³⁰removed: To our knowledge, there are only a few publications where the stability of the World Reference and Arosa Triads is analyzed. In these articles, and due to the large number of ozone measurements performed throughout day, the authors have calculated a representative value of all of them and, from it, the long-term stability of its triad has been analyzed (Fioletov et al., 2005; Stübi et al., 2017; Scarnato et al., 2010).

¹³¹removed: Fioletov et al. (2005) studied the long-term stability of the World Triad Reference in the period 1985 – 2003. In this work, the authors proposed to fit the measurements performed by each Brewer (

¹³²removed: 008, #014 and #015) to a 2nd grade polynomial:

[..¹³⁴]

[..¹³⁵]183 during the year 2011. The vertical lines represent situations which can produce a change in the behaviour of the instrument, while the horizontal line represents the operative *ETC* used to calculate the TOC (Eq.1). As it can be observed, the ETC changed twice, the first time by maintenance tasks (performed by IOS service in July 2011), and the second time due to changes in the Brewer configuration (to be more precise, changes in the so-called “Cal-Step” in August 2011). On the contrary, during the maintenance tasks (June 2011) or after UV calibration in our facilities (November 2011), the [..¹³⁶]

[..¹³⁷]

[..¹³⁸]ETC remained constant. Only the Langley's that satisfy the conditions indicated in Sect. 2.2 are used to calculate the weekly mean. Other examples of events that may affect the ETC can be found in the calibration campaign reports (Redondas et al., 2015; Redondas and Rodriguez-Franco, 2016).

[..¹³⁹]

[..¹⁴⁰]When a new *ETC* is given, the [..¹⁴¹]TOC calculated from it can be compared with the data obtained by other instrument with similar precision. This can be a [..¹⁴²]strategy to check if the new ETC is correct. At the RBCC-E Triad, this task is simple because the Brewers are constantly compared to each other, allowing to identify the exact moment when a Brewer needs a new calibration. In addition, the traceability between the RBCC-E and the [..¹⁴³]

[..¹⁴⁴][..¹⁴⁵][..¹⁴⁶]

[..¹⁴⁷]

¹³⁴removed: where O_3 are the ozone concentrations measured and $t - t_0$ corresponds to the difference between the time of the measurement and the solar noon. The independent coefficient A obtained through the adjustment is used as a representative ozone value of each instrument. The difference between this coefficient for each Brewer and the Triad mean represents the drifts of each instrument. The stability is studied from daily and monthly mean of these differences.

¹³⁵removed: Stübi et al. (2017) studied the long-term stability of the Arosa Triad in the period 1988 – 2015. In this study,

¹³⁶removed: authors considered that the Triad is the most appropriate reference for each day. Therefore, the measurements of the three Brewers are modeled as a 3rd grade polynomial dependent on time :

¹³⁸removed: where t_0 corresponds to the 12 UTC time. In this case, each Brewer is characterized by a shift Δ , which is the mean of the difference between the values measured and obtained from the fit, and a standard deviation σ . The standard deviation σ evaluates the dispersion of these differences. Both parameters are used to analyze the long-term stability of the Arosa Triad.

¹³⁹removed: In order to compare the long-term stability of the RBCC-E Triad with respect to the World Reference and Arosa Triads, both expressions are used to fit our measurements in this work. In this work, the time reference t_0 is the solar noon.

¹⁴⁰removed: Although Eqs. 8 and 9 are valid to model the behavior of ozone, it should be noted that

¹⁴¹removed: presence of outliers can affect the final value of the adjustment. Given this problem, calculating the daily mean of the measurements

¹⁴²removed: good strategy to avoid this inconvenient. In this work, and knowing that ozone presents a stable behaviour throughout the day at tropical latitudes,

¹⁴³removed: mean of all measurement of each Brewer was used to calculate a representative value for each Brewer .

¹⁴⁷removed: From these values, and following the procedure proposed by Fioletov et al. (2005), the long-term stability of our triad was also studied.

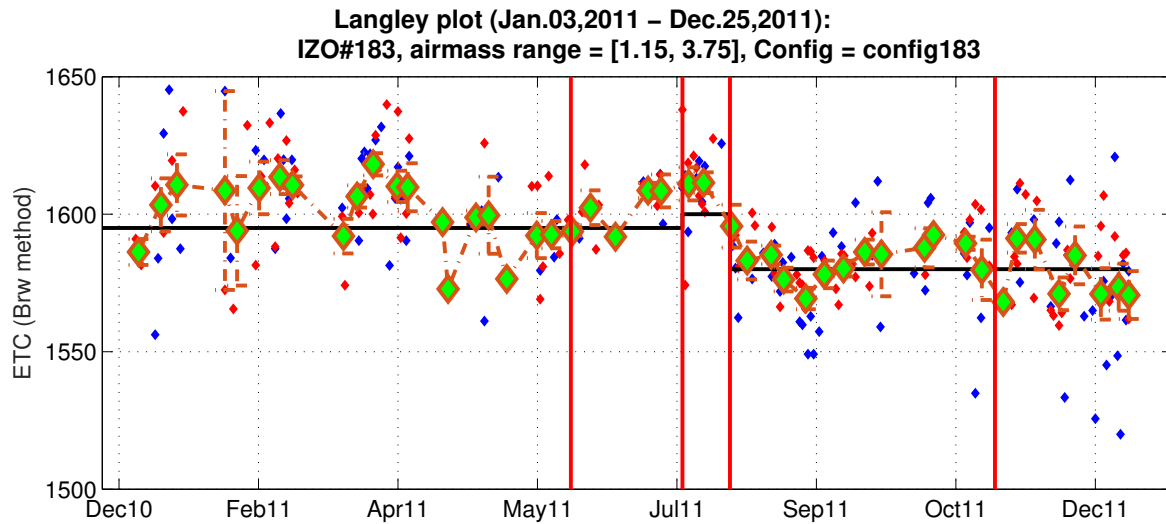


Figure 2. Operative ETC value for Brewer #183, after some events that could cause a change in the instrumental calibration during 2011. The red and blue dots denote daily ETC values calculated by the Langley method before and after solar noon, respectively. The diamond symbols and the error bars correspond to the ETC weekly mean and its standard deviation.

[..¹⁴⁸] World Reference Triads is checked in the calibration campaigns, organized by the RBCC-E, comparing the data of the travelling references: Brewers #[..¹⁴⁹]185 and #[..¹⁵⁰]017. The results of these comparisons are shown in the calibration campaign reports (Redondas et al., 2015; Redondas and Rodríguez-Franco, 2015; Redondas and Rodríguez-Franco, 2016).

5 2.1 [..¹⁵¹]

3 Ozone and Dataset selected

In order to summarize the history of the RBCC-E Brewers, Table 1 provides the total number of days and measurements performed by these instruments since they became operational at IZO and as long as the weather conditions allowed them to operate.

¹⁴⁸removed: Finally, the short-term, or intra-day, stability is also studied to identify the SZA ranges at which the differences between the measurements of

¹⁴⁹removed: 157, #183

¹⁵⁰removed: 185 are highest and lowest. This analysis was performed following the procedure proposed by Stübi et al. (2017), where the measurements are separated as a function of the SZA

¹⁵¹removed: Dataset selection criteria.

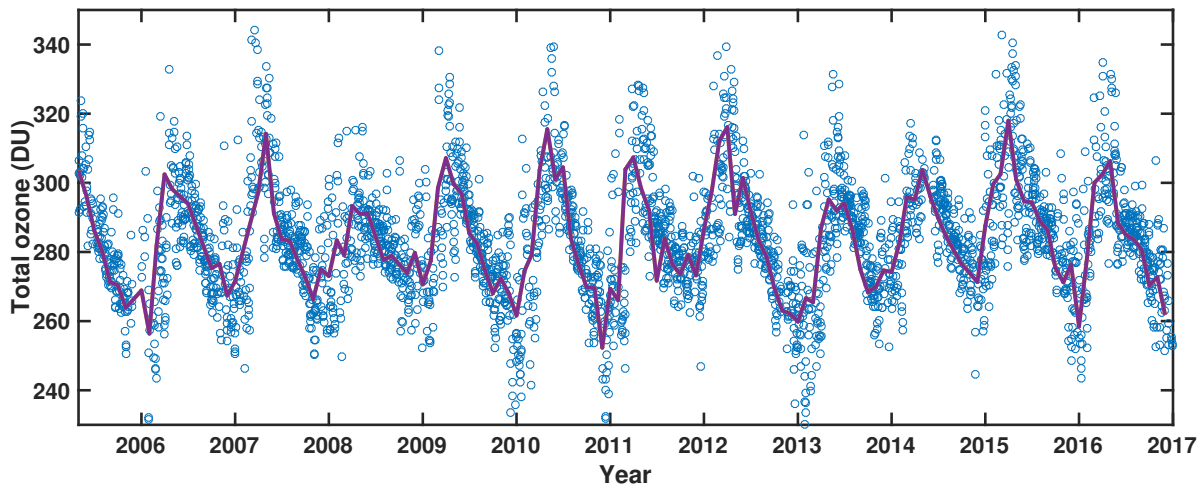


Figure 3. Time series of the ozone concentrations measured by Brewer #157 at Izaña Atmospheric Observatory, showing daily (dots) and monthly (line) means.

[..¹⁵²] Fig. 3 shows the daily and monthly TOC means measured by Brewer #¹⁵³ [¹⁵⁴][¹⁵⁵][¹⁵⁶][¹⁵⁷][¹⁵⁸][¹⁵⁹][¹⁶⁰][¹⁶¹][¹⁶²][¹⁶³]

[¹⁶⁴]157. The ozone presents an annual cycle with a sawtooth profile with maximum and minimum values in spring and autumn, respectively. Despite this annual behaviour, the ozone presents a lower daily variability as indicated by our measurements. This factor, together with a thermal inversion which produces an atmosphere free of anthropogenic pollutants and excellent weather conditions all the year, except for days with dust intrusions, explain why Izaña Atmospheric Observatory is an excellent location for a Brewer reference centre and, also, why the Langley-technique is used as calibration method.

In this work, the ozone datasets have been analyzed. The first dataset is obtained [¹⁶⁵]directly, after applying several conditions, listed next in order of application:

¹⁵²removed: Number of operational days and measurements since their setup for the Brewers of the RBCC-E Triad.

¹⁵³removed: 157

¹⁵⁴removed: Brewer #183

¹⁵⁵removed: Brewer #185

¹⁵⁶removed: Operational Days

¹⁵⁷removed: 3173

¹⁵⁸removed: 2740

¹⁵⁹removed: 2780

¹⁶⁰removed: Operational Measurements

¹⁶¹removed: 259534

¹⁶²removed: 204022

¹⁶³removed: 229201

¹⁶⁴removed: The stability of the RBCC-E Triad was evaluated from two datasets

¹⁶⁵removed: from the times series available for Brewers #157, #183 and #185,

1. Only include measurements performed at Izaña Atmospheric Observatory.
2. Remove days with problems clearly identified (wrong alignment, etc.).
3. Only the days where the three Brewers have performed measurements are considered in this work. Moreover, each Brewer must have more than 12 measurements, with a minimum of 4 before and after the solar noon (homogeneous distribution).
4. [..¹⁶⁶]

The second dataset is obtained with the same conditions but also imposing the condition that the measurements must be simultaneous [..¹⁶⁷]in time. A measurement made by a Brewer is considered simultaneous if [..¹⁶⁸]in a temporal window of five minutes there is other measurement made by the other two [..¹⁶⁹]brewers of the triad. Therefore, this second dataset can be considered a subset of the first. Table 2 gives a summary of both datasets. The entry “Evaluated Days” denotes the number of days used in each dataset to study the stability of the RBCC-E Triad. It is important to note that [..¹⁷⁰]Dataset 1 includes the measurements made in the period 2005-2016 while Dataset 2 only [..¹⁷¹]considers simultaneous measurements from 2010 onwards. Before 2010, a large part of the measurement time was focused on the UV spectrum, and, hence, there are fewer ozone direct sun measurements in the instrument schedule. This means that the likelihood of finding 12 simultaneous measurements between the three instruments is low, particularly in winter where the presence of clouds is [..¹⁷²]greater. After 2010, The RBCC-E started using the same synchronization schedule in their Brewers. These schedules take into account the sunrise and sunset times of each day [..¹⁷³]and the routines, introduced in it, are distributed in function of the solar zenith angle (SZA).

Table 1. Number of operational days and measurements since their setup for the Brewers of the RBCC-E Triad.

	Brewer #157	Brewer #183	Brewer #185
Operational Days	3173	2740	2780
Operational Measurements	259534	204022	229201

¹⁶⁶removed: The standard deviation of the measurements performed by each Brewer must be lower than 0.6. This is introduced to avoid days where the ozone presents an unusual behaviour.

¹⁶⁷removed: (condition 3 above)

¹⁶⁸removed: it is within 5 minutes of measurements performed

¹⁶⁹removed: Brewers of the RBCC-E Triad

¹⁷⁰removed: the

¹⁷¹removed: considered

¹⁷²removed: higher

¹⁷³removed: . Therefore, there is one for each day of the year. The routines

Table 2. Summary of the datasets used in this work.

	Dataset 1	Dataset 2
Evaluated Days	2073	1325
Period studied	2005–2016	2010–2016

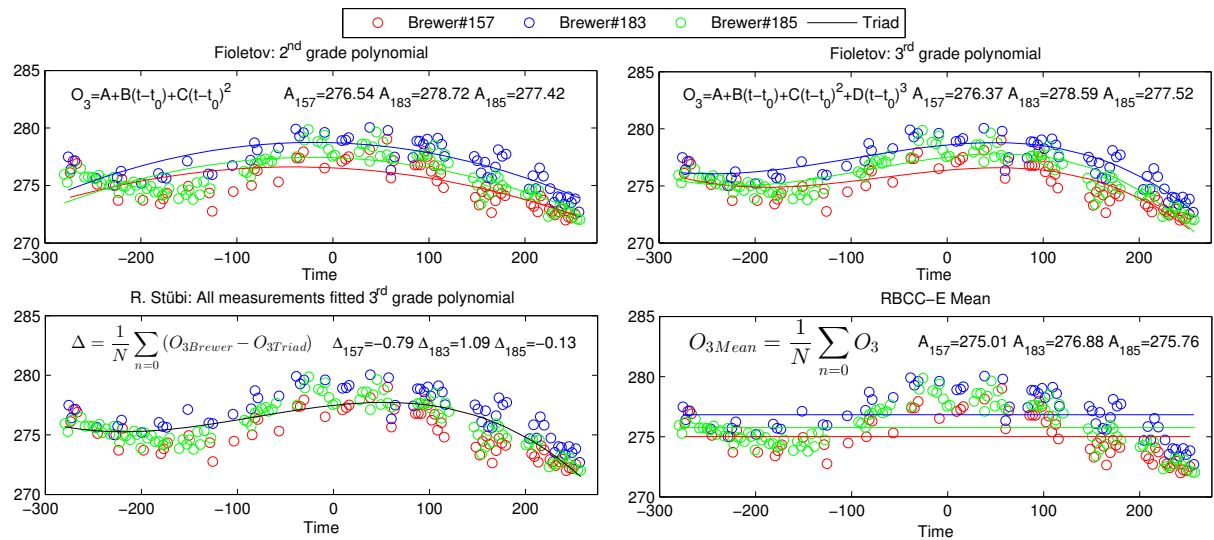


Figure 4. Method used to calculate the daily representative value. The ozone values plotted were measured the 16th November 2016.

4 Results and discussion.

4.1 [..¹⁷⁴]

[..¹⁷⁵]

The stability of the measurements carried out by the RBCC-E Triad was evaluated from the datasets described in Sect. [..¹⁷⁶]3. In the case of the long-term behaviour, it was studied using both datasets, while the short-term behaviour was analyzed using only Dataset 1. The results obtained are shown in statistical terms.

4.1 Representative value of the Total ozone column.

To our knowledge, there are only a few publications where the stability of the World Reference and Arosa Triads are analyzed. In these articles, and due to the large number of ozone measurements performed each day, the authors have

¹⁷⁴removed: Langley and ozone at RBCCE triad.
¹⁷⁵removed: As mentioned
¹⁷⁶removed: 2.2, the ETC can be used as a probe of the internal state of a Brewer. In the period 2005–2016, the ETC used

calculated a representative value of all of them and, from it, the long-term stability is analyzed (Fioletov et al., 2005; Scarnato et al., 2010; Stübi et al., 2017).

Fioletov et al. (2005) studied the long-term stability of the World Triad Reference in the period 1985 – 2003. In this work, the authors proposed to fit the measurements performed by each Brewer [..¹⁷⁷] (#008, #014 and #015) to a 2nd grade polynomial (see Fig.4):

$$O_3 = A + B \cdot (t - t_0) + C \cdot (t - t_0)^2 \quad (8)$$

where O_3 is the TOC measured and $t - t_0$ corresponds to the difference between the time of the measurement and the solar noon. The independent coefficient A obtained through the adjustment is used as a representative ozone value of each instrument. The difference between this coefficient for each Brewer and the Triad mean represents the daily drifts of each instrument. The stability is studied from the relative standard deviation of these differences. In this work, the [..¹⁷⁸] results using a 3rd grade polynomial have also been investigated, see Fig.4.

[..¹⁷⁹] Stübi et al. (2017) studied the long-term stability of the Arosa Triad in the period 1988 – 2015. In this study, the authors considered that the behaviour of the [..¹⁸⁰] Triad is the most appropriate reference for each day. Therefore, all the measurements made by the three Brewers are modeled as a 3rd grade polynomial dependent on time which represents to the [..¹⁸¹] Triad (see Fig.4):

$$O_3 = A + B \cdot (t - t_0) + C \cdot (t - t_0)^2 + D \cdot (t - t_0)^3 \quad (9)$$

where t_0 corresponds to the 12 UTC time. In this case, each Brewer can be characterized by a shift, Δ , which is the daily mean of the difference between the values measured and obtained from the fit and a standard deviation, σ , which evaluates the dispersion of these differences. Both parameters are used to [..¹⁸²] analyze the long-term stability of the Arosa Triad.

¹⁷⁷removed: has been modified several times. Different events can cause that a new calibration is necessary – e. g.

¹⁷⁸removed: replacement of components during maintenance tasks (micrometers, H. V. source, etc.) or by damages caused during bad weather situations (high winds, snowfall). Also, the ETC must be modified if a parameter of the Brewer configuration has been changed (dead time, temperature coefficients, filter attenuation, etc).

¹⁷⁹removed: As an example, Fig. 2 shows the operative value of the ETC for the Brewer #183 during the year 2011. The vertical lines represent situations which can produce a change in

¹⁸⁰removed: instrument, while the horizontal line represents the operative ETC used to calculate the TOC concentration (Eq.1). As it can be observed, the ETC was changed two times,

¹⁸¹removed: first time by maintenance tasks (performed by IOS service in July 2011), and the second one due to changes in the Brewer configuration (to be more precise, changes in the so-called “Cal-Step” in August 2011). On the contrary, during the maintenance tasks (June 2011) or after UV calibration in our facilities (November 2011), the ETC remained constant. Only the Langley’s that satisfy the conditions indicated in Sect. 2.2

¹⁸²removed: calculate the weekly mean. Other examples of events that may affect the ETC can be found in the calibration campaign reports (Redondas et al., 2015; Redondas and Rodriguez-Franco, 2016)

[..¹⁸³]In order to compare the long-term stability of the RBCC-E [..¹⁸⁴]

[..¹⁸⁵]

[..¹⁸⁶]

[..¹⁸⁷]Triad with respect to the [..¹⁸⁸]World Reference and Arosa Triads, both methods are used to fit our measurements.

5 However, in this work, the time reference t_0 is the solar noon.

[..¹⁸⁹]Although Eqs. 8 and 9 are valid to model the behaviour of ozone, it should be noted that the normal drifts by its continued operation of the instrument can affect the final value of the adjustment. Given this problem, calculating the daily mean of the measurements can be a good strategy to avoid this inconvenience. In this work, and noting by our measurements that ozone presents a reduced daily variability,see Fig.4, the mean of the all measurements, N, made by

10 each Brewer was used as a representative value.

$$A_M = \frac{\sum_i O_3}{N} \quad (10)$$

The difference between the value obtained for each Brewer and the Triad mean represents the drifts of each instrument. Although the median can be another possibility to study the behaviour of the [..¹⁹⁰]RBCC-E Triad, our experience suggests that the mean is robust. Moreover, since 2003, the mean has been used to detect when one of our Brewer loses
15 its calibration, therefore, it has been interesting to include it in this work. At this point, it is important to note that for the World Triad Reference as for the RBCC-E, the representative value of each instrument is calculated, directly, from their measurements. In contrast, in the Arosa Triad, the representative value of each instrument (denoted as shift Δ) is calculated with respect to the behaviour of the three instruments, obtained by adjusting to a polynomial of the third degree (see Fig.4).

¹⁸³removed: When a new ETC is given, the TOC concentration calculated from this new configuration can be compared with the values obtained by other instrument with similar resolution. This is the best strategy to check if the new ETC is the correct one and allows to identify the exact moment when a Brewer begins to have an irregular behaviour and needs a new calibration. At the RBCC-E this task is simple because the Brewers are constantly compared to each other. The tractability between the

¹⁸⁴removed: and the world Reference Triad during the calibration campaigns throughout the travelling references #185 and #017 which is a regular section of the calibration campaign reports.

¹⁸⁵removed: Operative ETC value for Brewer #183, after some events that could cause a change in the instrumental calibration during 2011. The dots in the figure denote ETC values calculated by the Langley method before and after solar noon each day. The diamond symbols and the error bars correspond to the ETC weekly mean and its standard deviation.

¹⁸⁶removed: Time series of the ozone concentrations measured by Brewer #157 at Izaña Atmospheric Observatory, showing daily (dots) and monthly (line) means.

¹⁸⁷removed: Fig. 3 shows the daily (circles) and monthly (line) TOC means measured by Brewer #157. As it can be observed, the ozone presents an annual cycle with a sawtooth profile with maximum and minimum values in spring and autumn, respectively. Despite this annual behaviour, the ozone is stable during the day, with a low standard deviation for

¹⁸⁸removed: recorded values. This factor, together with a thermal inversion which produces an atmosphere free of anthropogenic pollutants and excellent weather conditions all the year, explain why Izaña Atmospheric Observatory is an excellent location for a Brewer reference centre and, also, why the Langley technique is used as calibration method.

¹⁸⁹removed: In the next section it is shown that

¹⁹⁰removed: calibrations used for each Brewer and introduced after an event are correct, and then the long and short-term stability of the ozone values recorded by each Brewer is evaluated.

4.2 [..¹⁹¹]

[..¹⁹²]

4.1.1 Long-term stability[..¹⁹³]: daily averages

Following the procedure described by Fioletov et al. (2005) for the World [..¹⁹⁹]Triad Reference, Datasets 1 and 2 (see Sect. 3) were fitted by a 2nd [..²⁰⁰]and 3rd grade polynomial[..²⁰¹]. The distribution of the daily difference between the [..²⁰²]A value, see Eq.8, obtained for each Brewer with respect to the Triad mean are plotted in Fig. 5. Also, in this plot, the [..²⁰³]difference calculated from the daily mean of each instrument was included, see Eq.10.

It is important to take into account that the individual coefficients obtained for each Brewer, and also the Triad mean calculated from them, depend on the method used. As Fig.5 shows, the histograms that represent the results obtained after applying a polynomial fit are similar regardless of the dataset[..²⁰⁴]. This can be explained by the small daily variation of the ozone [..²⁰⁵]what causes the 2nd and 3rd grade polynomial fit to give very similar [..²⁰⁶]A coefficients, see Fig.4. In contrast, the histograms associated with the daily mean suggest that the differences between the brewers are less. This allows us to conclude that the method selected to evaluate the stability [..²⁰⁷]plays an important role, because the Brewer-Triad mean differences are directly associated with [..²⁰⁸]it. In this case, it may be more appropriate to use the daily mean to evaluate the RBCC-E Triad.

Regardless, independently of the method used, Fig. 5 shows that, for the great majority of days, the Brewers present less than 2 DU of difference with respect to the Triad mean [..²⁰⁹]which indicates a good agreement among themselves.

Using the same procedure to evaluate the long-term stability can be the best strategy to compare different triads to each other. In this sense, Fioletov et al. (2005) only reported [..²¹⁰]that the relative daily standard deviation of World triad reference is equal to 0.47%. This value [..²¹¹]was calculated as the mean of the relative standard [..²¹²]deviation of each brewer.

¹⁹¹removed: Stability of the RBCC-E Triad.

¹⁹²removed: The consistence of the measurements carried out by the RBCC-E Triadwas evaluated from the methods and datasets described in Sect. 3. In the case of the long-term stability, it was studied using both datasets, while the short-term stability was analyzed using only Dataset 1. The results obtained are shown in statistical terms.

¹⁹³removed: .

¹⁹⁹removed: triad

²⁰⁰removed: grade polynomial. In addition, in this work the results using a

²⁰¹removed: have also been investigated

²⁰²removed: A value

²⁰³removed: daily difference calculate from daily mean, A_M , of all measurements is included.

²⁰⁴removed: in contrast to the mean

²⁰⁵removed: at tropical latitudes. Therefore,

²⁰⁶removed: A coefficients. Therefore, the

²⁰⁷removed: of the measurements made by the Brewers

²⁰⁸removed: the selected method

²⁰⁹removed: . This result suggests that the Brewers of the RBCC-E Triad are in

²¹⁰removed: as daily data that the standard deviation mean of the Canadian Triad

²¹¹removed: is the average

²¹²removed: deviations of each Brewer

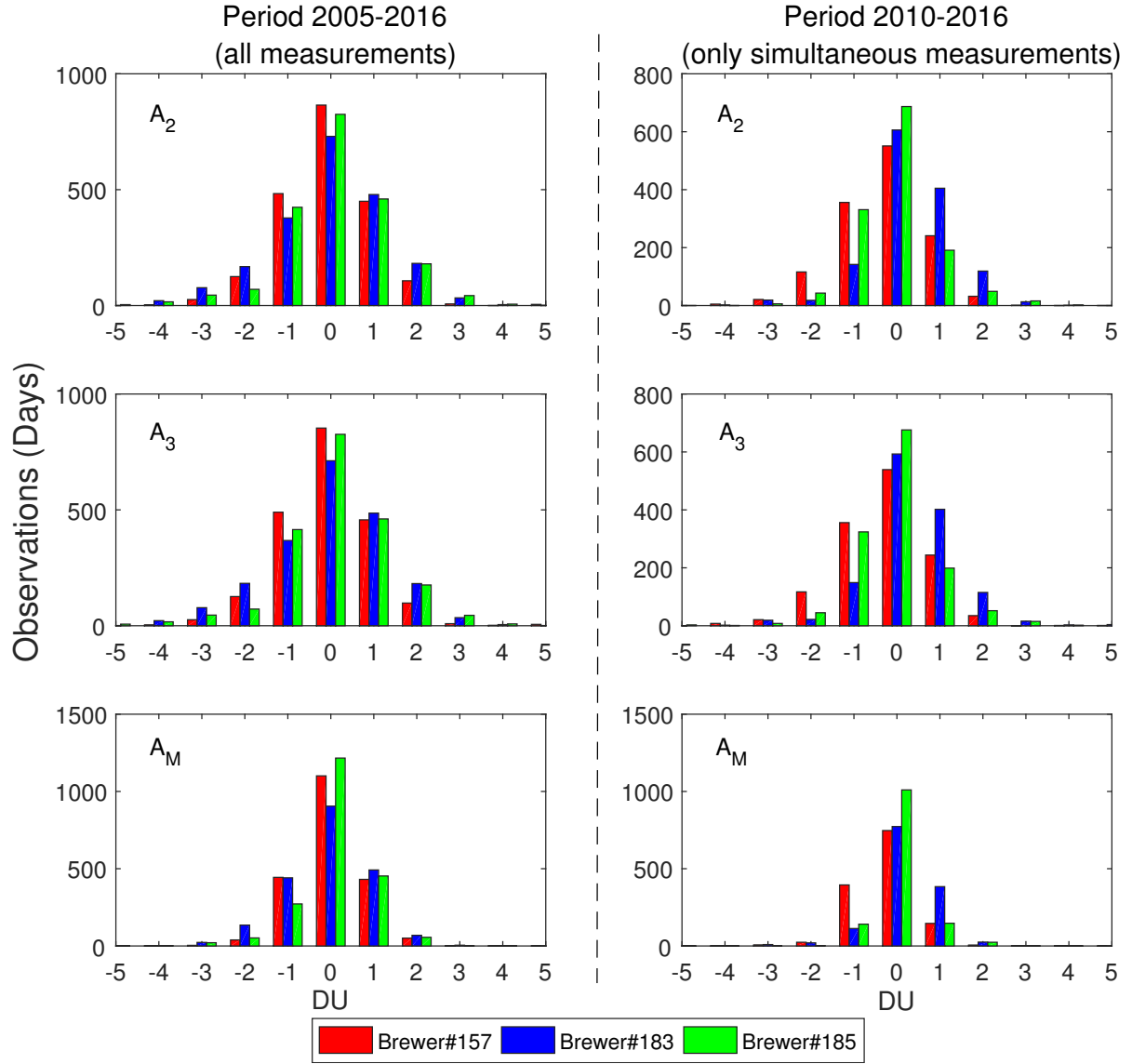


Figure 5. Daily difference of the ozone reference value A of each Brewer with respect to Triad. The values were obtained from the procedure proposed by Fioletov (World Triad Reference) and by daily mean (RBCC-E).

Table 3 contains the difference mean, calculated from the mean Brewer-Triad difference plotted in Fig. 5, and its standard deviation. The RBCC-E Triad presents a relative standard deviation mean equal to 0.41% ($\sigma_{157} = 0.362\%$, $\sigma_{183} = 0.453\%$ and $\sigma_{185} = 0.428\%$; see Table 3, Dataset 1, 4^{th} column). This result indicates that the dispersion of the measurements of the

²¹³removed: column $A_2(\%)$

Table 3. Absolute and relative values of the mean shift and the standard deviation.

Dataset 1						
	[.. ²¹⁶]2 nd grade polynomial (DU)	3 rd grade [.. ²¹⁷]polynomial (DU)	Brewer [.. ²¹⁸]Mean (DU)	2 nd grade [.. ²¹⁹]polynomial (%)	3 rd grade [.. ²²⁰]polynomial (%)	Brewer [.. ²²¹]Mean
Brewer #157	0.787 ± 1.04	0.796 ± 1.07	0.569 ± 0.744	0.276 ± 0.362	0.279 ± 0.372	0.1994 ± 0.1
Brewer #183	0.989 ± 1.29	1.01 ± 1.31	0.741 ± 0.956	[.. ²²²]0.349 ± 0.453	[.. ²²³]0.356 ± 0.463	[.. ²²⁴]0.26 ± 0
Brewer #185	0.89 ± 1.21	0.90 ± 1.23	0.56 ± 0.78	[.. ²²⁵]0.315 ± 0.428	[.. ²²⁶]0.32 ± 0.438	[.. ²²⁷]0.20 ± 0
Dataset 2						
	[.. ²²⁸]2 nd grade polynomial (DU)	3 rd grade [.. ²²⁹]polynomial (DU)	Brewer [.. ²³⁰]Mean (DU)	2 nd grade [.. ²³¹]polynomial (%)	3 rd grade [.. ²³²]polynomial (%)	Brewer [.. ²³³]Mean
Brewer #157	0.795 ± 1.00	[.. ²³⁴]0.82 ± 1.05	0.534 ± 0.661	0.278 ± 0.349	0.286 ± 0.368	0.186 ± 0.2
Brewer #183	0.747 ± 0.942	0.784 ± 1.02	0.547 ± 0.719	[.. ²³⁵]0.262 ± 0.331	[.. ²³⁶]0.275 ± 0.36	[.. ²³⁷]0.192 ± 0
Brewer #185	0.64 ± 0.87	0.67 ± 0.99	0.376 ± 0.526	[.. ²³⁸]0.227 ± 0.311	[.. ²³⁹]0.238 ± 0.333	[.. ²⁴⁰]0.133 ± 0

RBCC-E Brewers presents [..²¹⁴]a similar behaviour to those of the [..²¹⁵]World Triad Reference. Furthermore, the standard deviation values obtained confirm that the daily mean is the best method to evaluate the RBCC-E Triad.

In order to compare the daily behaviour of the Arosa and RBCC-E Triads, a 3rd grade polynomial was fitted to all the daily measurements made by [..²⁴¹]RBCC-E Brewers for Datasets 1 and 2. Then, for each Brewer its mean shift, Δ , and standard deviation, σ , were calculated [..²⁴²](see Sect. 3). The values obtained for the Dataset 1 are shown in Fig. 6. Because Brewer #183 was damaged by a storm and was inoperative between December 2005 and September 2006, the data plotted in that period were calculated from measurements of Brewers #157 and #185 only. Similarly, when Brewer #185 is away from IZO in calibration campaigns, the values plotted correspond to Brewers #157 and #183. Note that although these data were introduced in Fig. 6 to avoid gaps in the plot, they are not considered in the statistical study. Therefore, the dates evaluated correspond with the days when the full RBCC-E Triad is operative, and the criteria established in Sect. 3 are still used.

As [..²⁴³]can be observed in Fig. 6, the results obtained for all instruments in Dataset 1 show a (± 0.5) value for the mean shift. A similar result was obtained for Dataset 2 [..²⁴⁴], figure not shown [..²⁴⁵]. Contrary to the report in Stübi et al. (2017), in the present case the standard deviation does not show any seasonal component. Again, this result is explained by the [..²⁴⁶]low

²¹⁴removed: the same behaviour as

²¹⁵removed: Canadian Triad

²⁴¹removed: the

²⁴²removed: (Stübi et al., 2017).

²⁴³removed: it

²⁴⁴removed: (

²⁴⁵removed:)

²⁴⁶removed: almost constant value

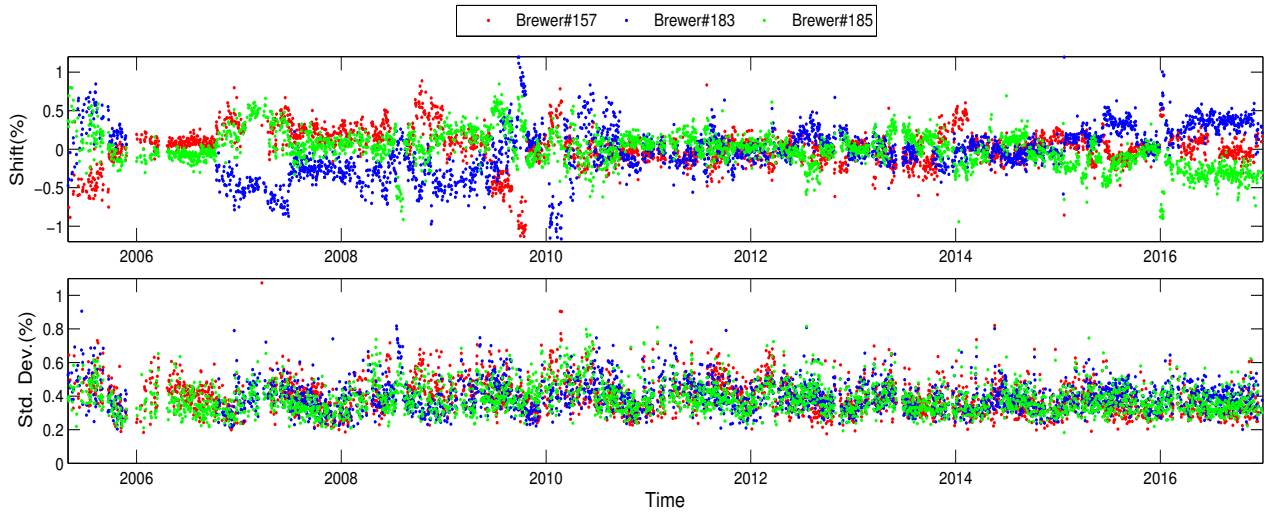


Figure 6. Time series of the mean shift Δ and the standard deviation σ , in terms relative to the TOC calculated from measurements performed by the three Brewers of the RBCC-E Triad (#157, #183 and #185) fitted with a 3rd grade polynomial

daily variability of the ozone at [..²⁴⁷]sub-tropical latitudes. For the Brewers of the RBCC-E Triad, the standard deviation is more influenced by any anomalous internal [..²⁴⁸]behaviour of the instruments. For middle latitudes, e.g. in Arosa, there is a larger daily variation in ozone and the standard deviation shows it.

Following Stübi et al. (2017), Table 4 shows the distribution of percentiles of the mean shift and the standard deviation values plotted in Fig. 6. [..²⁴⁹]The Brewers present a similar interpercentile range $P_{2.5} - P_{97.5}$, with a mean value close to 1.1%. This result is consistent with the standard deviation shown in Table 3 for the polynomial fits [..²⁵⁰]. In comparison with the Arosa Triad, only Brewer #040 shows a better [..²⁵¹]behaviour than the RBCC-E Brewers [..²⁵²]while their other Brewers (B#072, B#156) show similar values to [..²⁵³]ours.

4.1.2 Long-term stability: monthly averages

10 Although the histogram and the statistical parameters already presented suggest that the long-term stability of the RBCC-E [..³³⁴], Arosa and World Reference Triads are similar. It can be more interesting to [..³³⁵]present this study from the

²⁴⁷removed: tropical

²⁴⁸removed: behavior

²⁴⁹removed: All

²⁵⁰removed: , where the long-term stability of the Brewer-Triad mean was studied by the procedure proposed by Fioletov et al. (2005)

²⁵¹removed: behavior

²⁵²removed: . The other two Brewers of the Arosa Triad

²⁵³removed: those of the RBCC-E

³³⁴removed: Triad is similar to that of the World Reference and Arosa Triads , it

³³⁵removed: study the stability using

Table 4. Percentiles of the difference distribution (%) for the RBCC-E Triad.

Dataset 1		Shift Δ					Standard Deviation			
Brewer	$P_{2.5}$	P_{25}	Median	P_{75}	$P_{97.5}$	Brewer	$P_{2.5}$	P_{25}	Median	
#157	[²⁵⁴]-0.426	[²⁵⁵]-0.13	[²⁵⁶]0.017	[²⁵⁷]0.138	[²⁵⁸]0.464	#157	[²⁵⁹]0.25	[²⁶⁰]0.32	[²⁶¹]0.37	
#183	[²⁶⁴]-0.71	[²⁶⁵]-0.219	[²⁶⁶]-0.016	[²⁶⁷]0.191	[²⁶⁸]0.58	#183	[²⁶⁹]0.25	[²⁷⁰]0.327	[²⁷¹]0.37	
#185	[²⁷⁴]-0.54	[²⁷⁵]-0.099	[²⁷⁶]0.0155	[²⁷⁷]0.147	[²⁷⁸]0.51	#185	[²⁷⁹]0.24	[²⁸⁰]0.316	[²⁸¹]0.37	
Triad	[²⁸⁴]-0.599	[²⁸⁵]-0.14	[²⁸⁶]0.008	[²⁸⁷]0.156	[²⁸⁸]0.524	Triad	[²⁸⁹]0.248	[²⁹⁰]0.321	[²⁹¹]0.37	

Dataset 2		Shift Δ					Standard Deviation			
Brewer	$P_{2.5}$	P_{25}	Median	P_{75}	$P_{97.5}$	Brewer	$P_{2.5}$	P_{25}	Median	
#157	[²⁹⁴]-0.573	[²⁹⁵]-0.265	[²⁹⁶]-0.053	[²⁹⁷]0.084	[²⁹⁸]0.365	#157	[²⁹⁹]0.25	[³⁰⁰]0.33	[³⁰¹]0.38	
#183	[³⁰⁴]-0.475	[³⁰⁵]-0.093	[³⁰⁶]-0.037	[³⁰⁷]0.264	[³⁰⁸]0.559	#183	[³⁰⁹]0.265	[³¹⁰]0.349	[³¹¹]0.40	
#185	[³¹⁴]-0.341	[³¹⁵]-0.103	[³¹⁶]0.007	[³¹⁷]0.114	[³¹⁸]0.513	#185	[³¹⁹]0.247	[³²⁰]0.33	[³²¹]0.38	
Triad	[³²⁴]-0.503	[³²⁵]-0.136	[³²⁶]-0.001	[³²⁷]0.1386	[³²⁸]0.517	Triad	[³²⁹]0.242	[³³⁰]0.33	[³³¹]0.38	

monthly means. With this idea in mind, the [³³⁶]daily difference plotted in Fig. 5 and [³³⁷]the daily shift plotted in Fig.6 were monthly averaged. Fig.7 shows, in relative terms, the monthly values [³³⁸]for the period 2005–2016 (Dataset 1). The results confirm that the RBCC-E Triad has a good long-term [³³⁹]precision, regardless of the method selected. In order to compare with the World Triad Reference, Table 5 contains the relative standard deviation of the [³⁴⁰]difference between the representative value A of each brewer, calculated from the 2^{nd} grade polynomial fit, and the triad mean. As reported by Fioletov et al. (2005), the relative standard deviation of the 3-monthly mean for the World Triad Reference is 0.40%, 0.46% and 0.39% for Brewers #008, #014, and #015, respectively ([³⁴¹]0.42% in mean). The RBCC-E Brewers have lower [³⁴²]1-monthly and 3-monthly relative standard deviation. The ratio between 3-monthly values is 40% lower for the RBCC-E.

Furthermore, Stübi et al. (2017) reported that in the period 2004–2012 the Arosa Triad presented monthly [³⁴⁶]shift around at $\pm 0.4\%$ [³⁴⁷]while the RBCC-E are lower than [³⁴⁸] $\pm 0.3\%$.

³³⁶removed: values plotted in Figs

³³⁷removed: 6 were

³³⁸removed: of A_2 , A_3 , A_M and shift σ obtained for each Brewer in

³³⁹removed: stability, regardless

³⁴⁰removed: standard deviation obtained from the monthly values of the A_2 coefficients was calculated

³⁴¹removed: 0.4

³⁴²removed: monthly values 0.33% , 0.34% and 0.23% for Brewers

³⁴⁶removed: values lower than

³⁴⁷removed: . The corresponding monthly values for

³⁴⁸removed: $\pm 0.1\%$

Table 5. RBCC-E and World Reference Triads: Relative monthly standard deviation.

	RBCC-E		WORLD REFERENCE	
	1-Monthly	3-Monthly	3-Monthly	
Brewer #157 [.. ³⁴³]	0.33%	0.29%	Brewer #008	0.40%
Brewer #183 [.. ³⁴⁴]	0.34%	0.31%	Brewer #014	0.46%
Brewer #185 [.. ³⁴⁵]	0.23%	0.20%	Brewer #015	0.39%
Mean	0.33%	0.27%	0.42%	

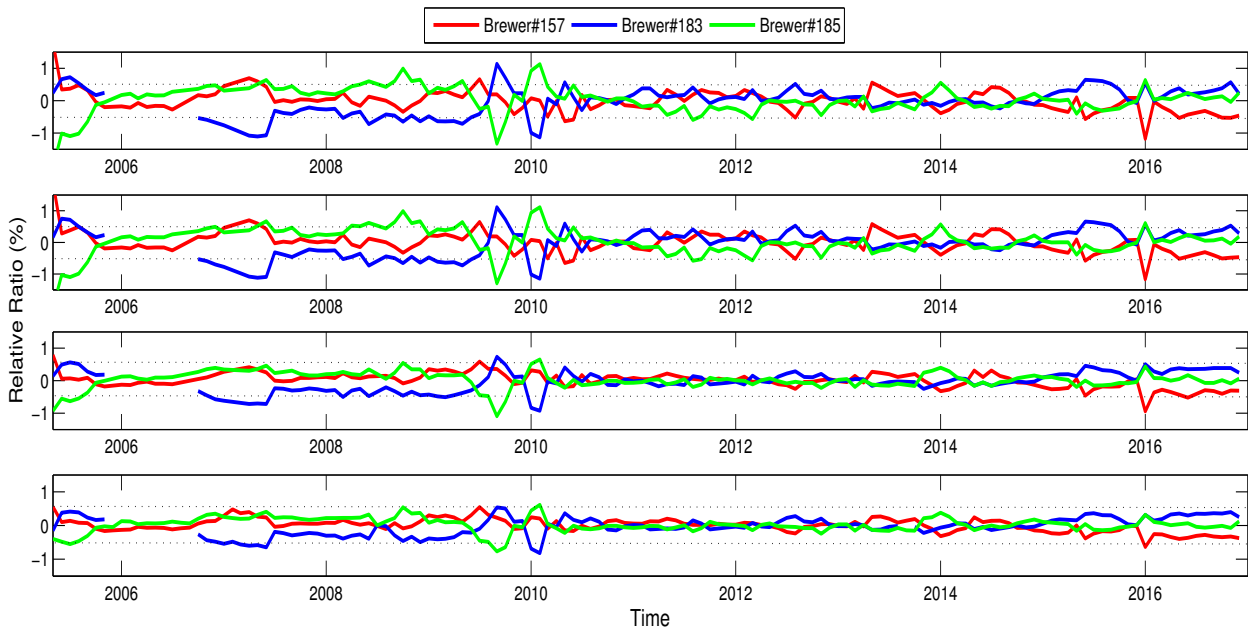


Figure 7. Relative ratio of the monthly values with respect to the Triad mean for [..³⁴⁹]the [..³⁵⁰]method proposed for World Triad Reference, [..³⁵¹]Fioletov et al. (2005), Daily Mean (RBCC-E) and [..³⁵²]Arosa Triad, [..³⁵³]Stübi et al. (2017)[..³⁵⁴]. The gap for the Brewer #183 data was caused by the tropical storm “Delta” which damaged the instrument. In 2010, the Brewer #183 had a problem with their micrometers

4.2 Short-term stability[..³⁵⁵]

Dataset 1 was used to study the short-term stability of the RBCC-E Triad, with a view to determine in which SZA range the consistence of the measurements is higher. The measurements made by the three Brewers every day were fitted by 3rd grade

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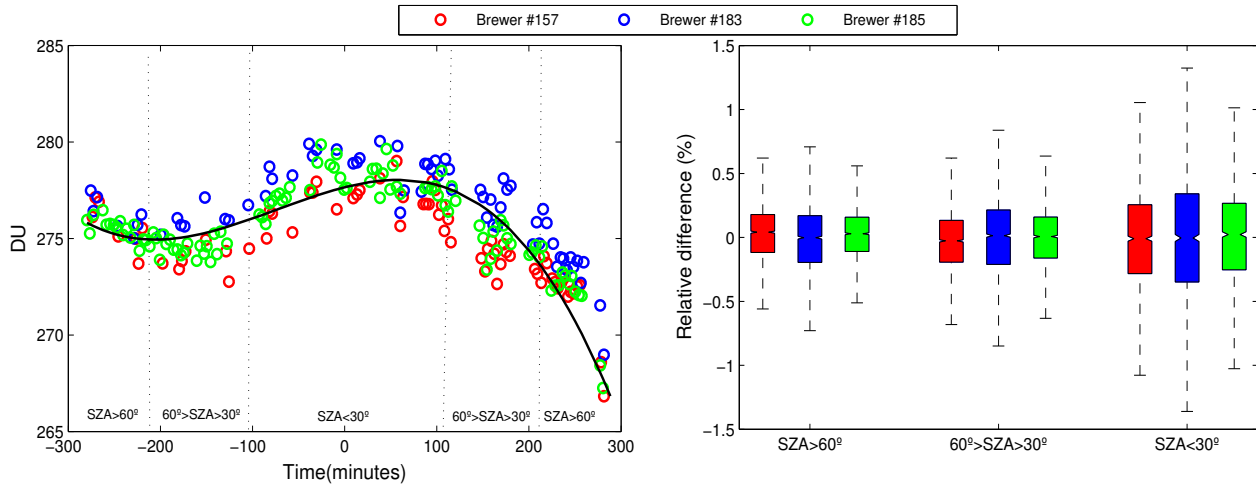


Figure 8. Experimental measurements and 3rd grade Triad fit (left) and relative difference by Brewer as function of the SZA (right).

polynomial as shown in Fig. 8. As previously commented, this polynomial represents the [\[..³⁵⁶\]](#)behaviour of the triad and allow [\[..³⁵⁷\]](#)obtain the TOC as a function of the time. Similarly to the previous study, each Brewer was characterized by a shift Δ . In this case, the [\[..³⁵⁸\]](#)data were divided as a function of the SZA. Different SZA ranges were checked, finding that the analysis can be reduced to just three broad ranges:

- 5 1. SZA > [\[..³⁵⁹\]](#)60° corresponds to the first and last ozone measurements of every day, when solar radiation presents a low intensity and high Rayleigh scattering.
2. SZA < [\[..³⁶⁰\]](#)30° corresponds to the measurement in the middle of the day, when the air mass is close to 1 and, hence, there is less Rayleigh scattering.
3. [\[..³⁶¹\]](#)60° < SZA < [\[..³⁶²\]](#)30°, the rest of ozone measurements.

- 10 In Fig. 8, the box-plot shows the statistical distribution of the mean shift calculated [\[..³⁶³\]](#)for the ranges selected. As can be observed, the greatest dispersion values are at low SZA [\[..³⁶⁴\]](#)which indicates at solar noon when more discrepancy can

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³⁶²removed: 30

³⁶³removed: in function on the SZA. As it

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be observed between the ³⁶⁵data recorded by the ³⁶⁶instruments. This result may seem ³⁶⁷surprising because in these conditions the solar radiation on the Earth's surface is maximum and the Rayleigh scattering is minimum ³⁶⁸but it can be easily explained from Eq. 1. In this expression, at low SZA the optical mass³⁶⁹, μ , is close to 1 and the ozone absorption α is a constant value. ³⁷⁰This implies that the denominator takes an almost constant value. Therefore, a small fluctuation (noise) associated with the MS9 values may affect significantly the ³⁷¹TOC recorded. In contrast, for the other ranges the denominator takes a significant value and the effect of the noise on MS9 is less, increasing the stability of the measurements. In addition, the Fig. 8 shows that the Brewer #183 presents the poorer results which can be explained if it is taken into account that this instrument was damaged in 2007, and during 2008 it had an irregular operation. Moreover, in 2010, it had a problem with their micrometers (Roozendael et al., 2013).

³⁷²

5 Conclusions

The consistency of ³⁷³TOC measurements made by the RBCC-E Triad has been studied in order to check its long-term stability ³⁷⁴to compare it with the values reported for the World Reference and Arosa Triads. With this idea in mind, the ³⁷⁵procedures used by these triads were reproduced ³⁷⁶in this work to analyze the RBCC-E data. From the method used to evaluate the World Triad Reference, the ³⁷⁷difference between the measurements made by each Brewer and the Triad mean present a relative daily standard deviation ³⁷⁸equal to $\sigma_{157} = 0.362\%$, $\sigma_{183} = 0.453\%$ and $\sigma_{185} = 0.428\%$ ³⁷⁹(mean $\sigma = 0.41\%$), lower than the reported value for the World Triad Reference (³⁸⁰ $\sigma = 0.47\%$). Using monthly averages ³⁸¹of these differences, the relative standard deviation reports values slightly lower than those obtained from the

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³⁷²removed: In conclusion, the other range where the best stability can be observed. In this range, the solar radiation, the Rayleigh scattering and the optical mass are not in their extremes values and , consequently, a minimum variation of these does not produce important variations in the ozone concentration measured by the Brewer.

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³⁷⁴removed: and compare with the behavior

³⁷⁵removed: methods used to study the stability of

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Table 6. Summary of the three studies comparing of the relative standard deviation of the World Triad Reference, Arosa and RBCC-E Triads

	daily	monthly	3-monthly
World Triad Reference	0.42	-	0.47
Arosa Triad	0.5	0.36	-
RBCC-E	0.41	0.37	0.27

The standard deviation calculated from procedure of Fioletov et al. (2005) and Stübi et al. (2017) are not equal but they are similar enough to compare the three triads. In this table, the values introduced for the RBCC-E were obtained from the procedure used by Fioletov et al. (2005) for the World Reference Triad.

daily data. ³⁸²]In addition, applying the procedure used to study the Arosa Triad, the ³⁸³]RBCC-E Triad ³⁸⁴]presents a similar interpercentile range $P_{2.5} - P_{97.5}$ ³⁸⁵], with a value close to 1.1%, similar to those reported for the Arosa ³⁸⁶]Triad, except in the case of Brewer #040. ³⁸⁷]However, the monthly means are better for the RBCC-E Triad. ³⁸⁸]Despite these differences, the values reported for each Triads are fairly similar, see Table 6, which ensures the traceability of the

5 ozone measurements all around the world.

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³⁸²removed: Applying
³⁸³removed: Brewers of the
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³⁸⁵removed: interpercentiles for their mean shift which are
³⁸⁶removed: Brewers
³⁸⁷removed: The
³⁸⁸removed: The values obtained for the different

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