March 26, 2018

Response letter to Referee #3 on the manuscript "Monte Carlo method for determining uncertainty of total ozone derived from direct solar irradiance spectra: Application to Izaña results"

Authors: Anna Vaskuri, Petri Kärhä, Luca Egli, Julian Gröbner, and Erkki Ikonen

Article reference: amt-2017-403

Authors: We thank Anonymous Referee #3 for the valuable comments that helped us in improving the manuscript. We have included below our detailed responses to all comments.

Specific comments

"P.4. Table 1. – 3. The table for Uncertainties including the fraction of correlation need further explaining, especially since the issue of correlation is introduced much later in section 4. If these uncertainties have been published in that form earlier, quotation would be helpful in the figure caption. If not, these tables should be moved to section 4 prior to table 4 or in combination with table 4 since the explaining is done on p.15"

> Authors: We agree, and move Tables 1, 2 and 3 to Section 4. Most of the uncertainties are estimated for this manuscript or obtained from personal communication, and they have not been published elsewhere. We include a new paragraph in Section 4 to explain how the uncertainties are obtained:
>
> "The uncertainties due to radiometric calibration include factors such as the uncertainty of the standard lamp used, and the additional uncertainty due to noise and alignment. QASUME has been validated using various methods, thus the uncertainty due to calibration is low (Hülsen et al. (2016)). For QASUME and BTS, we assume the correlations to be equally distributed between full correlation, unfavourable correlation, and random correlation (Kärhä et al. (2018)). Spectra measured with AVODOR are significantly noisier, thus half of the uncertainty is associated to the random component. Values for instability of the calibration lamp are based on long-term monitoring. The lamp irradiances have been noted to gradually drop with no significant wavelength structure within the wavelength region concerned. Non-linearity values are estimations of the operators of the devices. Non-linearity is typically manifested so that the responsivity of the device changes gradually from high readings to low readings. This can cause significant change in the TOC values, thus we assume the correlation type to be unfavourable. Uncertainties due to device stability and temperature dependence are based on long-term monitoring. The changes have been found to be independent on wavelength in the region concerned, thus full correlation is assumed. Noise is the average standard deviation of typical measurements at noon over the wavelength region concerned. The wavelength scales of the devices have been checked using emission lines of gas discharge lamps. The uncertainty values given are the estimated standard deviations of the possible remaining errors after corrections. Wavelength error can introduce a significant change in TOC, because it introduces an error in the form of the derivative of the spectral irradiance. Thus, unfavourable correlation is assumed. Most of the uncertainty components are slightly wavelength dependent but to simplify

simulations, average uncertainty values are used over the wavelength range between 300 nm and 340 nm."

A reference is updated:

Kärhä P., Vaskuri A., Pulli T., and Ikonen E.: Key comparison CCPR-K1.a as an interlaboratory comparison of correlated color temperature, J. Phys.: Conf. Ser., 972, 012012, 2018. doi:10.1088/1742-6596/972/1/012012

"L4-8. Why is uncertainty of radiometric calibration of AVODOR so much higher than for the other instruments? Just because of low SNR in the UV region?"

Authors: Yes, the uncertainty is higher mainly because of the noise. AVODOR response is noisier and less stable as compared with QASUME and BTS.

"The stated uncertainties given in the tables are valid for the whole spectral range of the instrument? I would expect uncertainties related to radiometric calibration and measurement noise to be wavelength dependent. Or are these stated values the upper limit of uncertainties? In the following text, there is a lot discussion about straylight effects. However, there is no explicit uncertainty component related to straylight or straylight correction?"

Authors: It is true that the uncertainties and noise levels are slightly dependent on the wavelength. We simplified the simulations by using average values. A sentence on this is included to the text in Section 4: "Most of the uncertainty components are slightly wavelength dependent but to simplify simulations, average uncertainty values are used over the wavelength range between 300 nm and 340 nm."

Stray light level is difficult to estimate, and to calculate its effect on the TOC uncertainties, we would need the stray light matrix for each instrument (Nevas et al. (2014)) that we do not have. A more practical way of obtaining its effect is to compare the results with a device that does not suffer from stray light. In this case, QASUME is significantly better with respect to stray light than the two array spectroradiometers, BTS and AVODOR. We include discussion about this in the paper.

In the case of array spectroradiometers, the stray light and baseline noise severely affect the TOC analysis at large zenith angles when the fitting is performed with the relative least squares method. This can be seen in Fig. 1R as an inverse U-shape of TOC values determined from BTS and AVODOR spectra when the relative least squares minimisation

$$S = \sum_{i=1}^{n} w(\lambda_i)[E_s(\lambda_i) - E(\lambda_i)]^2 \quad (1R)$$

with $w(\lambda) = E(\lambda)^{-2}$ is used. If the least squares method is modified so that absolute residuals ($w(\lambda) = 1$) are minimised, the inverse U-shape of TOC results at large zenith angles diminishes. With the absolute least squares fitting, the TOC values at noon are 2 DU lower as compared with those estimated using the relative least squares fitting. We include this analysis and Fig. 1R in the revised manuscript. This issue is discussed in more detail in our response to Referee #2.

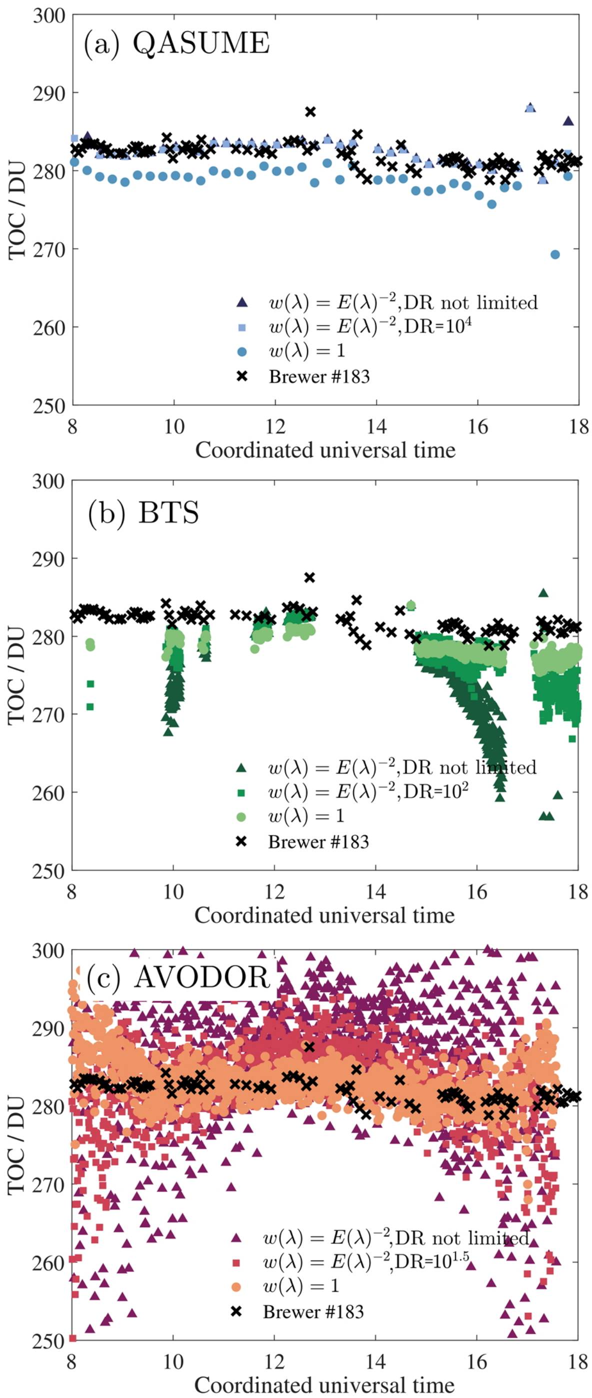

Figure 1R. TOC values estimated using different weighting in least squares minimisation and using Ångström AOD model for QASUME (a), BTS (b) and AVODOR (c). TOC values for Brewer #183 are plotted as black crosses for comparison. The colour codes and the associated figure legends denote the weighting used and the dynamic range DR used.

Reference:

Nevas S., Gröbner J., Egli L., and Blumthaler M.: Stray light correction of array spectroradiometers for solar UV measurements, Appl. Opt., 53, 4313–4319, 2014.

"P. 7, L15 The least square fitting might lead to local minima instead of the global minimum. How is that accounted for? Especially if AOD and TOC are both fitting parameters."

Authors: We performed the least squares fitting using a Matlab function 'lsqnonlin'. We set optimoptions(@lsqnonlin, 'Algorithm', 'trust-region-reflective', 'MaxFunEvals', 1000, 'MaxIter', 1000, 'StepTolerance', 1e-10, 'OptimalityTolerance', 1e-10, 'FunctionTolerance', 1e-10). In our case, one of these tolerances was met by less than 50 iterations when the initial guess values were set to TOC = 200, β = 0, and c = 1.

We included an offset factor c as free fitting parameter in front of $E_{\text{ext}}(\lambda)$ as suggested by Referee #2. After this change, we have three parameters (TOC, β, c) to be fitted. We also set $\beta \geq 0$ using the bound constraints of 'lsqnonlin' function, as aerosols attenuate solar UV spectrum.

To our understanding, global minimum should be achieved quite easily with two or three free parameters, but of course, it depends on algorithms used and the data set to be fitted. We tested the robustness of our retrieval by varying the initial guess values of TOC, β, and c. We varied the initial TOC value between 10 DU – 700 DU, the initial guess value of β between 0 – 0.5, and the initial guess value of c between 0 – 100. Using the initial guess values within such ranges, the free parameters always converged to the same final values and they were independent on the initial guess values. We include text on these tests in the AMT Discussion manuscript on page 7 after line 16.

"P.9, L20 "In this paper, we do this for all components, where the mechanism of contributing to the uncertainty of TOC is known." I guess these components are those with correlation "full" and "random"? This could be specified here."

Authors: Actually, the text refers to those components, where fractions for correlations are not listed in Table 4. Instead, they are labelled as (a) – (d). We clarify this by including a sentence before line 5 on page 14:

"For components (a) – (d) in Table 4, the mechanism of contributing to the uncertainty of TOC is known. We know the standard uncertainty of the O_3 layer altitude of 26 km to be u = 0.5 km, so we vary the altitude accordingly and note the variance of the resulting TOC."

"P.11, Figure 4 That figure is a bit confusing. For underlining the statement on full, unfavorable and random correlation the display of one colored graph is sufficient. The additional information gained from the u=5% and u=2.5% graph, as well as the black solid lines is not explained in the plain text and incomprehensible explained in the figure caption."

Authors: All those curves were intended to show the scalability of the model, but we agree that they were not sufficiently explained in the text. To avoid confusion, we replace Fig. 4 of our AMT Discussion manuscript with Fig. 2R shown below. The new Fig. 4 includes the values obtained by varying the spectral irradiance (circles) and also the values obtained by varying the ozone absorption cross section (crosses). The uncertainty behaves differently if the parameter to be analysed is in front of the equation as a multiplier (such as spectral irradiance) or in the exponent as a direct multiplier to TOC (such as the O_3 cross section). We include a new sentence on page 10, line 16, of the AMT Discussion manuscript as:

"The uncertainty in TOC arising from the spectral deviation in $\alpha_{O_3}(\lambda, T_{\text{eff}})$ is plotted as crosses in Fig. 4 as a function of increasing N."

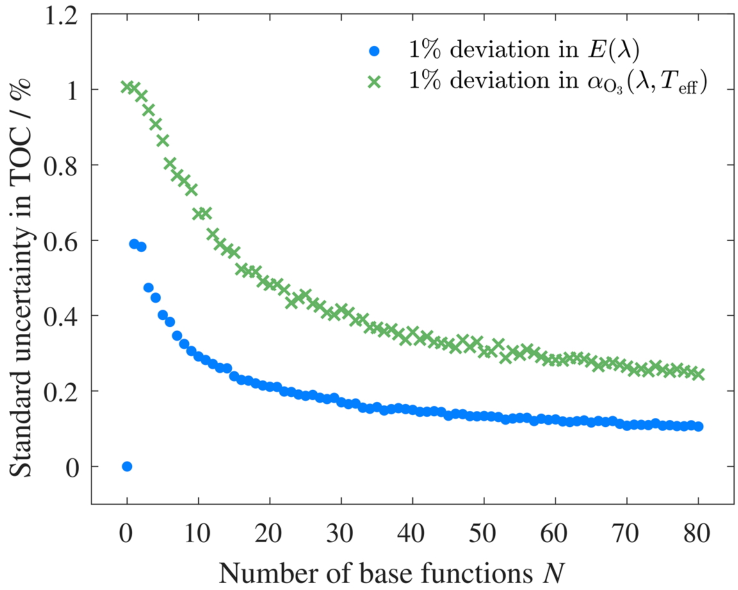

Figure 2R. Standard uncertainties of TOC at local noon as a function of the order of complexity N for QASUME spectroradiometer, with 1% standard deviation in spectral irradiance $E(\lambda)$ plotted as circles, and 1% standard deviation in ozone absorption cross-section $\alpha_{O_3}(\lambda, T_{eff})$ plotted as crosses.

Additional notes by the authors

As we modified the retrieval algorithm by including a new offset factor c to compensate for full spectral deviations, the results as compared to the AMT Discussion paper will slightly change. Please, see separate response letter addressed to Referee #2 for full details.