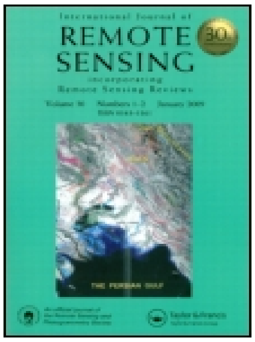

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Total ozone column measurements using an ultraviolet multi-filter radiometer

P.I. Raptis [a]*, S. Kazadzis [a,b], K. Eleftheratos[c], P. Kosmopoulos [a], V. Amiridis [d], C. Helmis[e], and C. Zerefos [f]

[a]*Institute for Environmental Research and Sustainable Development (IERSD), National Observatory of Athens (NOA), 15236 Athens, Greece;* [b]*Physikalisch-Meteorologisches Observatorium Davos, World Radiation Center, Davos, Switzerland;* [c]*Faculty of Geology and Geoenvironment, University of Athens, Athens, Greece;* [d]*Institute for Astronomy, Astrophysics, Space Applications and Remote Sensing (IAASARS), National Observatory of Athens (NOA), Athens, Greece;* [e]*Faculty of Physics, Departments of Environmental Physics and Meteorology, University of Athens, Athens, Greece;* [f]*Biomedical Research Foundation of the Academy of Athens, Athens, Greece*

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We have developed and used a method to retrieve total ozone column (TOC), from Ultraviolet Multi-filter Rotating Shadowband Radiometer (UVMFR) measurements in combination with radiative transfer model calculations. Look-up tables of ratios of the direct solar irradiance at (DI) 305 and 325nm in terms of TOC, solar zenith angle, and aerosol optical depth (AOD) have been constructed and compared with TOC retrievals estimated directly from UVMFR irradiance measurements. Sensitivity analysis of the influence of AOD on the calculated TOC has been investigated and found to be 1 Dobson unit per 0.1 change in AOD. We also examined the impact of ozone effective temperature on the TOC retrieval and found that it leads to a 0.9% change in TOC per K. UVMFR direct irradiance measurements in Athens, Greece, during the period July 2009–May 2014 were used to create a time series of high-temporal-frequency measurements (1 min for cloud-less conditions) of TOC, which facilitated an analysis of the diurnal variation of TOC. Comparison of the TOC retrievals from the UVMFR with co-located and synchronous daily TOC retrievals from a Brewer MKIV spectrophotometer showed very good agreement (correlation coefficient 0.98). Daily TOC retrievals from the UVMFR were within ±3% compared with the ones measured by the Ozone Monitoring Instrument overpasses on board the Aura satellite.

1. Introduction

Ozone is the most important absorber of ultraviolet (UV) radiation in the Earth's atmosphere, and has become a focus of great scientific and public interest over the past few decades due to growing awareness of the effects of UV on human health. Towards the end of the twentieth century, a significant reduction of stratospheric ozone was observed by various studies, both at the global scale (e.g. Fioletov 2008; Mäder et al. 2007) and also locally, e.g. over Greece (Varotsos, Kondratyev, and Cracknell 2000; Zerefos 2002). After the ratification of the Montreal Protocol on 1 January 1989, the concentration of ozone-depleting substances worldwide has reduced (WMO 2006); however, local exceptions to this trend are the subject of active research. For example, extremely low ozone values were observed in the Arctic during

*Corresponding author. Email: piraptis@meteo.noa.gr

early 2011 (e.g. Varotsos, Cracknell, and Tzanis 2012). Monitoring stratospheric ozone is crucial for detecting other local perturbations such as these and for analysing future trends, in particular the influence of ozone concentrations on incoming UV irradiance and climate change. Importantly, over northern mid-latitudes, a decline of ≈ 3.5% has been recorded in UV radiation in the recent years relative to the 1964–1980 mean value (Bais et al. 2014).

For monitoring stratospheric ozone, Dobson and Brewer spectrophotometers are the primary ground-based instruments in use. They have been in operation now for several decades and provide long (decadal) time series of spectral UV irradiance measurements. Through well-established techniques and empirical calculations (e.g. Redondas et al. 2014), they also provide total ozone column (TOC) retrievals. The World Meteorological Organization (WMO) has initiated actions such as the Ozone Mapping Center (http://lap.physics.auth.gr/ozoness2/) in order to unify such measurements and provide near-real-time TOC for the Northern hemisphere. Although ground-based measurements provide long multi-decadal time series at high temporal resolution, their spatial coverage of the planet is low, especially over oceans and for locations near the equator. On the other hand, satellite instruments such as the Ozone Monitoring Instrument (OMI) (Levelt et al. 2006) have been providing total ozone measurements globally since 2004, but at a rate of only 1–2 measurements per day. Portable multi-filter radiometers help fill in data gaps in the global spatio-temporal record by providing high temporal resolution measurements. In particular, the Ultraviolet Multi-filter rotating shadowband Radiometer (UVMFR) is an instrument designed to measure total and diffuse irradiance and to calculate the direct solar irradiance (DI) with high accuracy and frequency. Thirty-six UVMFRs are currently deployed by the United States Division of Agriculture (USDA, http://uvb.nrel.colostate.edu/UVB/uvb_network.jsf) UV-B monitoring and research network to monitor UV irradiance nationwide. In addition to its portability, the main advantages of this instrument are the automatic calibration procedures and the low operational cost. Compared to Brewer spectrophotometers, low effort is needed with UVMFRs for quality assurance and quality control (Bigelow and Slusser 2000; Slusser et al. 2005).

Gao et al. (2001) have suggested a spectral method for calculating TOC using measurements at four wavelengths from a UVMFR and showed that daily TOC values agreed with Brewer measurements to within 1.4%. Slusser et al. (1999) retrieved daily TOC values from UVMFR measurements using a look-up table (LUT) generated from a multiple-scattering radiative transfer code and validated them against Brewer and Dobson-derived TOC data. Despite using only a relatively small data set (four and five months of daily values, respectively), both studies concurred and showed good agreement between UVMFR and spectrophotometer TOC values. In addition, Tree and Slusser (2004) compared five months of UVMFR recordings at Mauna Loa, Hawaii, to TOMS satellite retrievals (one satellite overpass per day) and also found a good agreement but with a small and systematic underestimation of the TOC.

Here, based on the approaches of Gao et al. (2001) and Slusser et al. (1999) we have developed a simple method using DI measurements at two wavelengths (305 and 325 nm) and the radiative transfer model (RTM) LUTs to retrieve high-accuracy TOC estimates. The method is applied to five years of high-frequency UVMFR measurements to retrieve TOC at a complex (in terms of aerosol load) site in the centre of Athens, Greece. We examine the effect of aerosol optical depth (AOD) and ozone effective temperature on our method and introduce corrections that improve the results. Finally, we have compared this method's results with synchronous and co-located Brewer and OMI retrievals TOC.

2. Instruments and models

The period of measurements analysed is from July 2009 to May 2014 at the Athens ground-based Atmospheric Remote Sensing Station (ARSS), which has been in continuous operation to monitor ground irradiance levels and aerosol loadings over the Greek capital (Amiridis et al. 2009; http://apcg.space.noa.gr/index.php?option=112&client=1&langid=2). ARSS is located on the roof of the Biomedical Research Foundation of the Academy of Athens (BRFAA) (37° 54′ N, 23° 48′ E, 130 m above sea-level) near the city centre and 10 km from the sea (Gerasopoulos et al. 2011). In addition to the UVMFR a Brewer spectrophotometer (part of BRFAA), which has been providing total ozone measurements since 2003 (Zerefos and Eleftheratos 2007), has been operated in parallel. Next, we present a brief description of the operation of the two devices and data consistency checks are described following that.

The UVMFR (Yankee Environmental Systems, Inc) measures total and diffuse horizontal irradiance (DHI) in the UV part of the solar spectrum. Measurements are performed centred at seven wavelengths (300, 305, 311, 317, 325, 332, and 368 nm) with a 2 nm nominal full width at half maximum bandwidth. Signals in all channels are recorded every 10 s simultaneously by different photodiode detectors passing through a single Lambertian diffuser made of Teflon, and 1 min average values are stored. The DI component is calculated at the same time by deducting the measured components. UVMFR measurements are corrected using dark signal and angular response corrections (Krotkov et al. 2005). The corrected direct UVMFR measurements are used to calculate AOD (at 368 nm) through frequent Langley calibrations and comparisons with a Cimel (Cimel Electronique S.A.S) Sun-photometer that is operating at ARSS. More details on Cimel operation and measurements can be found at Holben et al. (1998). In this work, to ensure the consistency of the ratio of the DI at 305 nm and 325 nm, Langley calibrations at low AOD (<0.1) conditions have been used. Global (total) irradiance measurements from the UVMFR were used to distinguish cloud-free conditions for each of the 1 min measurements. Clouds are detectable in the measured UVMFR global irradiance (at 368 nm) since they cause larger variability than aerosols. To distinguish between cloudy and cloud-free conditions, we applied an updated version of the method of Gröbner and Kerr (2001), which is based on a comparison of the measured DI with RTM calculations in cloud-free conditions. More details about this quality control of the UVMFR measurements are presented in the next section in the context of Brewer spectrophotometer measurements. The Brewer single monochromator (Brewer 001) performs direct Sun measurements at five nominal wavelengths, namely 306.3, 310.1, 313.5, 316.8, and 320.0 nm. TOC is calculated from these spectral measurements by the differential absorption retrieval method (Staehelin et al. 2003). In more details:

$$X = M_{S11} = (M_{S9} - B_1)/(A_1 \times M_2), \tag{1}$$

where X is TOC and

$$M_{S9} = M_{S5} - 0.5M_{S6} - 1.7M_{S7}, \tag{2}$$

and M_{S5}, M_{S6}, and M_{S7} are the measurements of the intensities at the different wavelengths (ratios of wavelengths pairs used in the Brewer total ozone algorithm: M_{S4}: 306.3/316.8; M_{S5}: 310.1/316.8; M_{S6}: 313.5:316.8; M_{S7}: 320.1:316.8). B_1 is the extraterrestrial constant for the wavelengths used for ozone measurements and A_1 is the differential ozone absorption coefficient for the ozone measurements determined by a linear combination of ozone absorption coefficients of different wavelengths selected by the slit mask for ozone measurements (see below for calibration). M_2 is the optical air mass, determined by

$$M_2 = \sec(\arcsin((R/(R+Z)) \times \sin(\theta))), \tag{3}$$

where R = 6,371.009 km is the Earth's radius, Z = 22 km is the ozone layer height, and θ is the solar zenith angle (SZA). B_1 and A_1 are instrumental constants that are determined by comparisons with a standard instrument and checked or updated by the inter-comparison with the traveling standard instrument. Lamp tests are performed every day as part of the recommended programme of automatic measurements, which can be adapted by the operators (Staehelin et al. 2003). The Brewer 001 spectroradiometer is calibrated regularly by means of the travelling standard Brewer 017. The last two calibrations were performed in September 2010 and October 2013 by the International Ozone Service Inc.

In addition to the ground-based measurements, TOC data from the OMIs on board the Aura satellite were also analysed for the study period. OMI is a nadir-viewing UV/visible solar backscatter spectrometer on board the Aura satellite. OMI TOC is co-located overpass data with respect to the ARSS data and has a synchronization window of 60 min and satellite-station spatial distances lower than 50 km. A detailed description of the OMI instrument, some procedures of data processing, quality control/quality assurance procedures, calibration, and characterization can be found in Veefkind et al. (2006). The theoretical basis of the OMI ozone product algorithm for deriving the TOC from spectral scattered radiances can be found in Bhartia and Wellemeyer (2002).

In parallel, we have used the libRadtran radiation code (Mayer and Kylling 2005) in order to simulate DI ratios from the UVMFR by performing a grid of runs whose basic input parameters include the θ, AOD, TOC, and absorption ozone cross section and whose outputs comprise high-resolution DI spectra with a spectral resolution of 0.01 nm. For each run, we have used a constant aerosol profile (US Standard Atmosphere 1976) and a single scattering albedo of 0.9. To accurately simulate the UVMFR direct Sun measurements, retrieved DI scans were weighted with the spectral response of the UVMFR instruments (for 305 and 325 nm). LUTs were then produced using the following relation between the calculated DI ratio (305:325 nm) and the model input parameters:

$$I_{305} : I_{325} = f(\theta, X, \tau_{\text{aerosol}}). \tag{4}$$

In Figure 1, we present an example of the variation of the DI ratio from the LUT as a function of varying θ, and X for the aerosol-free case (AOD = 0). From the LUT and the quality-controlled data set of DI measurements, we constructed a primary TOC data set for the study period.

3. TOC retrieval

3.1. Sensitivity to aerosols and ozone effective temperature

We studied the aerosol effect on the retrieved TOC by calculating the AOD at 368 nm (this wavelength is preferable because ozone absorption is negligible) from UVMFR irradiance measurements using the Beer–Lambert law:

$$\tau_{368\text{aerosol}} = 1/\mu(\ln I_{\text{o }368}/\ln I_{368}) - \tau_{\text{ray}368}, \tag{5}$$

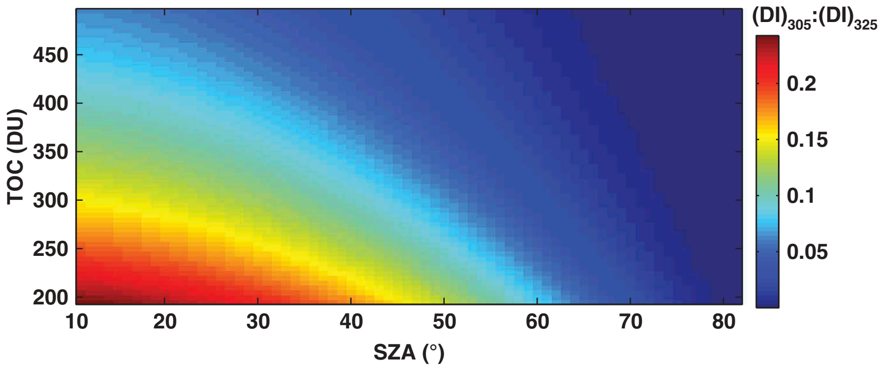

Figure 1. The variation of DI ratio (305:325) as a function of TOC and SZA (for AOD = 0) as calculated by RTM simulations.

where μ is the air mass factor, τ_{ray368} is the Rayleigh scattering optical depth at 368 nm that is calculated (Bodhaine et al. 1999), and $\tau_{368aerosol}$ is AOD at 368 nm. To investigate the sensitivity of the TOC retrieval to AOD, we performed RTM runs for different $\tau_{368aerosol}$ values in the range 0–1.2 with a 0.05 step size. We then recalculated the TOC data set by interpolating the corresponding AOD LUTs. Figure 2 presents the difference between TOC retrievals obtained with and without $\tau_{368aerosol}$ for the entire study period, which can be seen to be strongly linear (R^2 = 0.98). Averaging differences in 0.05 bins, a change in TOC of the order of 1 (±0.4) Dobson unit (DU) is observed for each 0.1 change in $\tau_{368aerosol}$. It is evident that for environments having high AOD variability such as the city of Athens (Kazadzis et al. 2012), the AOD correction presented here is essential for improving the accuracy of the TOC retrieval process. The potential of using synchronous UVMFR data to retrieve AOD is a great advantage of this method.

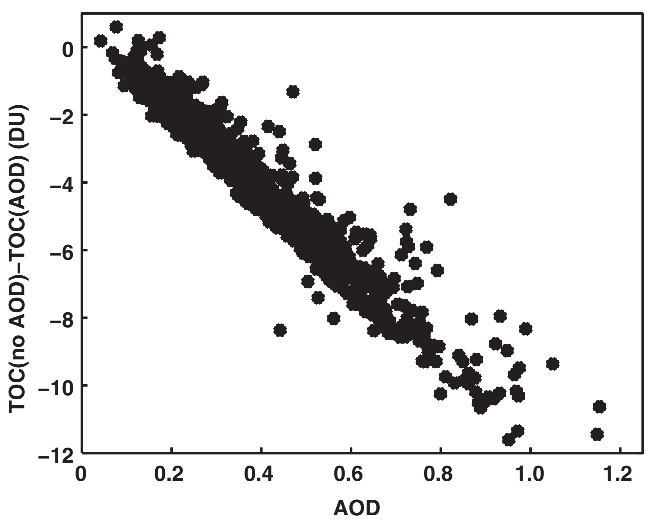

Figure 2. Bias of TOC calculated with and without aerosols, with respect to AOD at 368 nm.

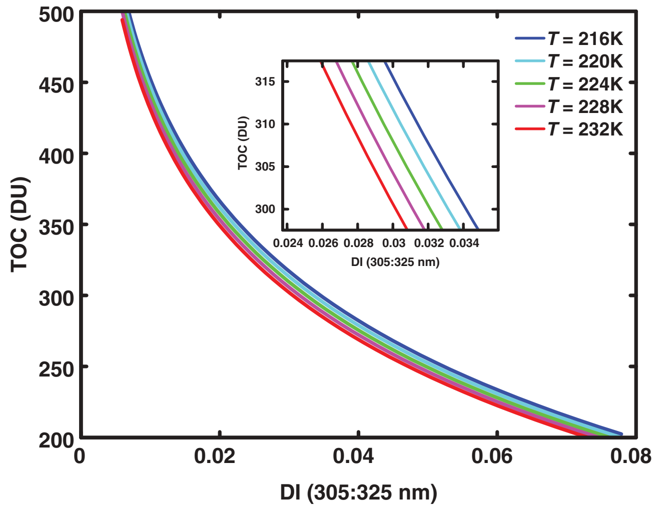

Figure 3. TOC as a function of the ratio of DI (305:325 nm) for Bass–Paur cross sections at different temperatures.

3.2. *Sensitivity to ozone effective temperature*

For the case of sensitivity of TOC retrieval to ozone effective temperature, we used the ozone absorption coefficients provided by Paur and Bass (1985) using different temperatures, as inputs to the libRadtran RTM. These coefficients are temperature-dependent, and ignoring them has been shown to lead to a seasonal error (Redondas et al. 2014) on TOC retrieval. Recent studies have also calculated the effect of temperature variation on ground-based measurements of TOC (Redondas et al. 2014; Fragkos et al. 2013). The most accurate approach requires ozone sonde data to calculate the ozone effective temperature and then applying the absorption coefficients. To provide a stand-alone and simpler method for retrieving the TOC, we proceeded as follows. The yearly variability of mid-latitude stratospheric temperature is around 16 K and the European Centre for Medium-Range Weather Forecasts (ECMWF) provides a mean value of 224 K over Greece (Parrish et al. 2013). This allows us to calculate ozone cross sections for the range of temperatures 216 K–232 K. Figure 3 shows the TOC values calculated at $\theta = 60°$ and $\tau_{368\text{aerosol}} = 0.5$ for different temperatures using this approach. We compared the TOC at 224 K to all tables for the same θ, and $\tau_{368\text{aerosol}}$, and a change in the calculated TOC of 0.5% per K was found in the temperature range of interest. Then, a correction to the calculated TOC data set was introduced using climatological values of stratospheric temperature.

4. Retrievals comparison

For validation of our results, we used the synchronous Brewer TOC retrievals described in the previous section. Since UVMFR measurements have a much higher measurement frequency than Brewer ozone measurements, interesting daily features could be identified. Figure 4(*a*), for instance, shows the daily TOC variability features captured. Such information is crucial when using the TOC column for the calculation of other parameters such as the UV index where its daily variability is required. Diurnal stratospheric ozone variation has been found to exhibit an afternoon peak at mid-latitudes related to the

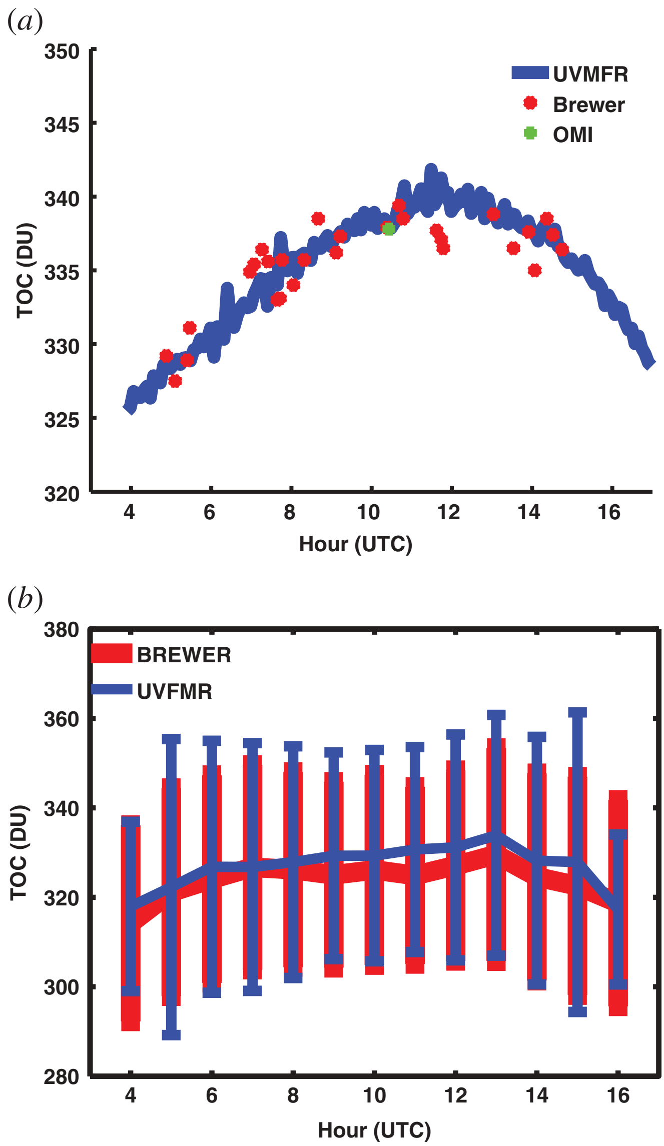

Figure 4. *(a)* Brewer, UVMFR, and OMI overpass TOC data on 17 June 2011. *(b)* Average hourly TOC values for both instruments, recorded during the month of June during 2011.

formation of tropospheric ozone near the Earth's surface at populated urban locations (e.g. Antón et al. 2010). Figure 4(*b*) shows the mean hourly values for both instruments during the month of June where an afternoon peak is visible in both data sets despite the high standard deviation calculated for both instruments. The physical explanation for this diurnal pattern is likely caused by the daily variability of photochemical processes related to ozone formation and destruction in the lower troposphere, especially for urban areas. According to Antón et al. (2010), these diurnal fluctuations in tropospheric ozone could explain part of the diurnal TOC variations (between 20% and 70% depending on the mixing layer height).

Table 1. Comparative statistics for TOC (Pearson product–moment correlation coefficient) and the total ozone ratio (mean, standard deviation σ) between the UVMFR and Brewer instruments retrievals for each OMI overpass data.

	Correlation coefficient (r)	Mean ratio	σ
UVMFR–Brewer	0.98	1.00	0.03
UVMFR–OMI	0.93	1.03	0.04
Brewer–OMI	0.96	0.99	0.03

The comparison was further assessed by studying the UVMFR/Brewer TOC ratios and the correlation coefficient between the two data sets. Results are presented in Table 1. We find a strong positive correlation between UVMFR and Brewer coincident measurements (r = 0.98) and with a mean total ozone ratio equalling 1.00 with a standard deviation of 0.03 95% confidence interval at ±0.002. Comparison with coincident OMI satellite retrievals also revealed strong positive correlations but with a small overestimation of the total ozone with the UVMFR compared with OMI, and a slight underestimation in the case of the Brewer instrument.

First we compared all of the values retrieved from UVMFR with the corresponding synchronous Brewer retrievals. For each Brewer measurement, we averaged 5 (min) UVMFR recordings. Measurements for SZA higher than 70° have not been used in the study in order to avoid uncertainties associated with the non-ideal angular response of the UVMFR instrument. We studied 24,723 cases of cloudless-sky synchronous measurements, presented in Figure 5, from which we determined a coefficient R^2 of 0.94. The average ratio is 1.024, with a standard deviation of 0.034. In addition, daily TOC values from the UVMFR and the Brewer have been calculated and are superimposed in Figure 5.

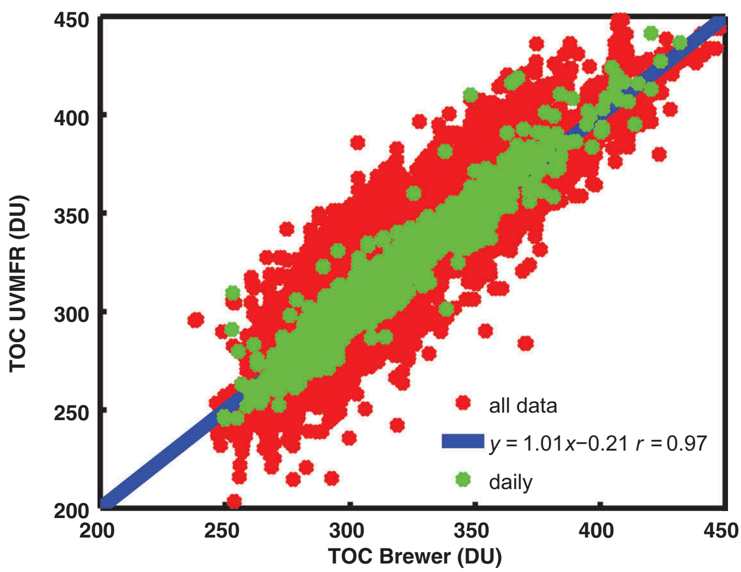

Figure 5. Scatterplot of all synchronous and quality assured values of Brewer and UVMFR TOC retrievals, in red and in green, all and daily values accordingly. Linear regression for daily values with r = 0.97.

Table 2. Comparative statistics for TOC ratio (mean, standard deviation σ) between the UVMFR and Brewer instruments retrievals for each OMI overpass data per month.

	<TOC UVMFR/ Brewer>	σ TOC UVMFR/ Brewer	<TOC UVMFR/ OMI>	σ TOC UVMFR/ OMI
January	1.02	0.04	1.02	0.03
February	1.00	0.03	0.98	0.02
March	0.99	0.03	0.98	0.03
April	0.98	0.02	0.98	0.04
May	1.02	0.03	1.03	0.06
June	1.04	0.04	1.06	0.05
July	1.02	0.03	1.04	0.05
August	1.03	0.05	1.04	0.05
September	1.02	0.03	1.03	0.03
October	1.00	0.02	1.01	0.02
November	0.99	0.02	1.00	0.02
December	1.00	0.04	1.01	0.03

The mean median and standard deviations of the TOC ratio UVMFR/Brewer and UVMFR/OMI are presented in Table 2 for each month.

In addition, in Figure 6, we show the relative frequency histograms for the TOC ratio among UVMFR and Brewer for all synchronous values (blue) and daily values (red). Difference in the skewness is observed, revealing a slight UVMFR overestimation when using all values statistics. Using a *t*-test distribution we calculated 95% confidence intervals as 1.0085 ± 0.0003 for all values and 1.0075 ± 0.0020 for daily values. The main cause of this behaviour is the largest scattering of >1 values, which are mainly found in the summer months when more measurements are available.

In our aim to investigate the error sources linked with the TOC retrieval, we show the UVMFR/brewer ratio with respect to SZA in Figure 7. It appears that both the ratio and the scatter of values are independent of the SZA. A slight overestimation of the UVMFR retrievals appears at around 45°. Overall, it can be seen that the errors are within ±3.5%

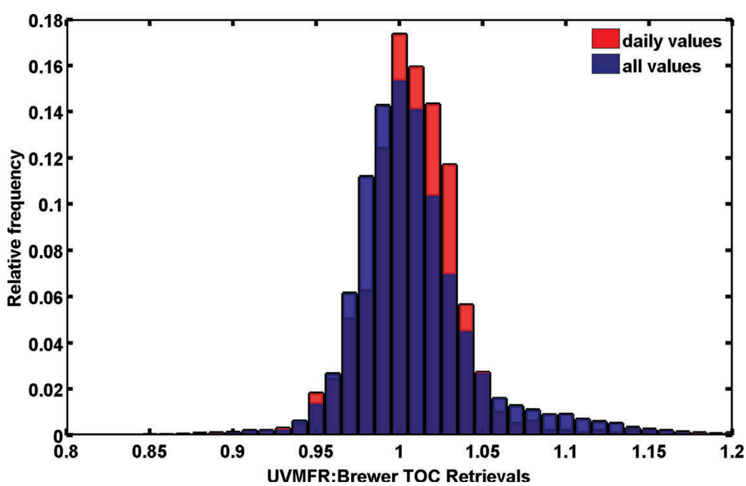

Figure 6. Relative frequency histogram of all TOC ratio (UVFMR:Brewer) values and daily values correspondingly.

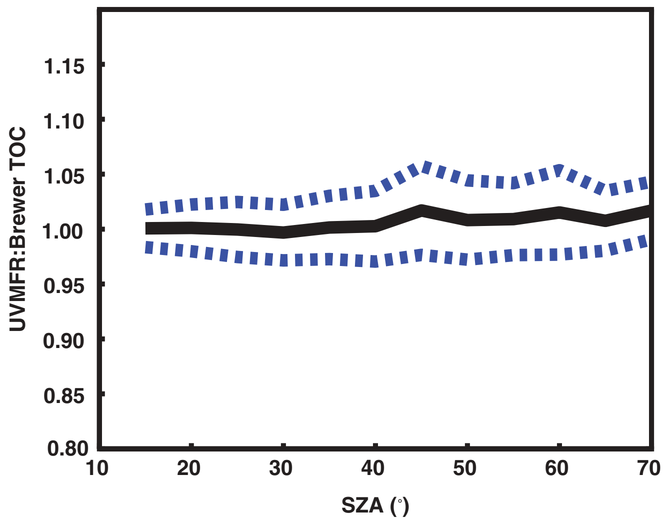

Figure 7. UVMFR:Brewer TOC all values ratio, averaged at 5° bins. Blue dashed lines indicate ±1 one standard deviation.

for all SZAs. The non-SZA-dependent ratios reveal the ability of the UVMFR to simulate the DI accurately and using the presented methodology to simulate TOC for SZA up to 70°. Examining the ratio among the UVMFR and Brewer time series, there appears a small remaining seasonality of the order of 3%; whereas during winter months UVMFR underestimates, it overestimates during the summer months. This difference suggests that despite the fact that seasonal correction is applied, a seasonal dependence remains on the calculated TOC.

To compare OMI-based and ground-based TOC measurements, we have produced UVMFR and Brewer daily values of TOC averaging measurements in a 2 hour window around OMI overpass (Figures 8 and 9). The annual variability of the mainly stratospheric

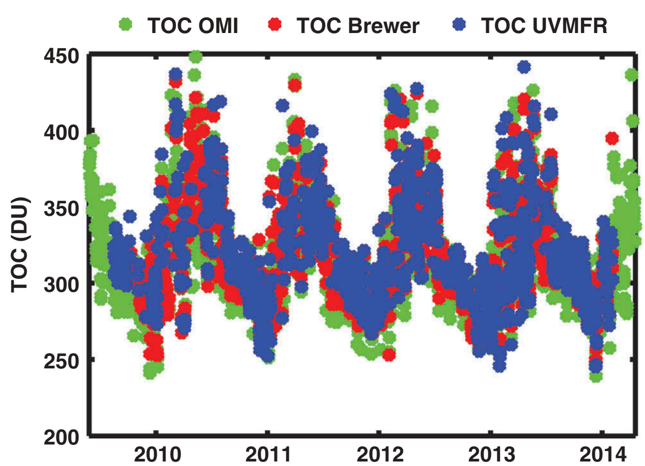

Figure 8. Daily TOC retrievals from OMI, Brewer, and UVMFR measurements over the period 2009–2014 in Athens, Greece.

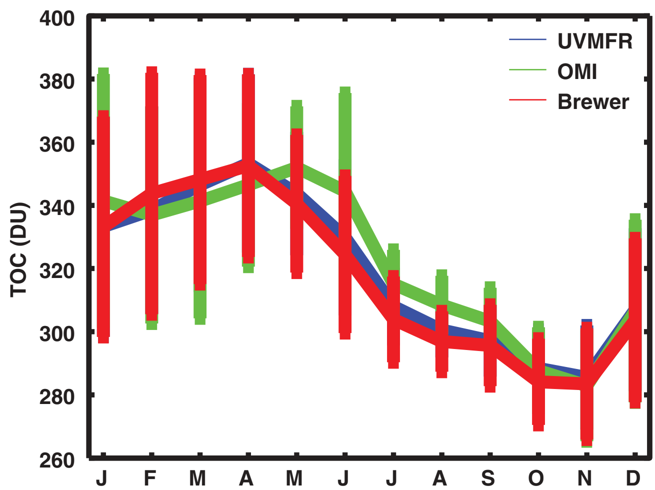

Figure 9. Monthly average TOC retrieved by Brewer and UVMFR accordingly, and the corresponding OMI overpass means, 2009–2014.

TOC is captured from all instruments. Hence, this approach can provide a satisfactory representation of the atmospheric TOC state. The results are presented in Figure 8 alongside OMI retrievals. Data presented here capture a span of 5 years (2009–2014) of continuous ozone measurements from the two surface-based instruments. All three methods show the same ozone-related seasonal cycle over Athens, with maximum values during April–May and minimum during October–November, which is better visualized in Figure 9. This figure reveals a slight overestimation of OMI around summer months and an underestimation in spring, which is caused by the different ozone cross sections used for the TOC retrieval. Balis et al. (2007) had found average biases among the TOC retrievals and Brewer measurements of OMI to be less than 3%. Our results show a good agreement between the Brewer and the satellite total ozone, which is of the order of 2.7% with a standard deviation of 4%. Similar results have been found comparing TOC from OMI and UVMFR.

5. Conclusions

A simple method using RTM LUTs and UVMFR measurements at two wavelengths can be used to calculate TOC and it provides results comparable to other standard methods. Previously, Gao et al. (2001) and Slusser et al. (1999) had introduced the potential of using the UVMFR instruments for TOC measurements. In this work, we provide a sensitivity study and a validation of such UVMFR-based algorithms, introducing improvements based on AOD, and ozone effective temperature described effects on the retrieval algorithm. In addition to the previous works using TOC-UVMFR retrievals, this is the first work using a long-term (5 years) time series of TOC data. This is crucial to explore further the limitations of such a retrieval, especially linked with the long-term stability of the UVMFR instruments and their use for monitoring purposes.

The main advantages of the use of such a method include the low cost and relatively easier maintenance of the UVMFR compared with Brewer or Dobson TOC measuring spectroradiometers, the potential of high-frequency retrievals, and the ability to calculate synchronous AOD needed for improving the TOC retrievals. We analysed a 5 year UVMFR time series and compared it with collocated Brewer retrievals in Athens, Greece, and found a correlation in the order of 98%, mean ratio of synchronous values at 1, and a standard deviation of 0.003. We have shown that neglecting the AOD variations introduces an error of 1 DU per 0.1 change in AOD. We constructed and used LUTs that included SZA, AOD, and DI ratios of 305 and 325 nm. UVMFR retrievals slightly overestimate TOC in the summer and underestimate during winter, compared with the Brewer TOC retrievals. We investigated this seasonality and found that using the Bass–Paur ozone absorption coefficient should influence TOC by 0.5% per K. We used climatological values of ozone effective temperature to apply a correction on the retrieval. Moreover, we compared to OMI product and found differences in the order of ±3%. UVMFR irradiance measurements can be used to investigate diurnal variations of TOC.

The method could be easily adopted by any UVMFR instrument operating worldwide. Such initiatives and methodologies can be used to increase the low geographical coverage of current TOC instrumentation. Especially in the tropics where only few instruments are deployed, such initiatives could be a useful tool for TOC-related future networks. Comparing the difference on the cost and the maintenance needed for UVMFR and standard Brewer instruments, the use of UVMFR instrumentations together with developed algorithms such as the one presented in this work could be a step towards filling such surface-based TOC monitoring gaps.

Disclosure statement

No potential conflict of interest was reported by the authors.

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ORCID

P.I. Raptis http://orcid.org/0000-0002-4221-992X
S. Kazadzis http://orcid.org/0000-0002-8624-8247
P. Kosmopoulos http://orcid.org/0000-0001-9641-4141
V. Amiridis http://orcid.org/0000-0002-1544-7812
C. Zerefos http://orcid.org/0000-0002-4795-523X

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