# Ozone Determination by GUV 2511 Ultraviolet Irradiation Measurements at Stara Zagora

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Accepted: 17 February 2017

***Abstract*** A Ground-based Ultraviolet Radiometer (GUV) 2511was installed at Stara Zagora, Bulgaria, in February 2015. The GUV 2511 instrument was designed to measure the downwelling global solar irradiances at UV wavelengths 305, 313, 320, 340, 380, 395 nm and the irradiance at the wavelength interval from 400 to 700 nm in the visible range. The instrument allows a realistic estimate of the total column ozone (TCO) in the atmosphere, the evaluation of the UV-index and the retrieval of the cloud optical thickness. This study presents the methodology of the TCO retrieval from measurements of the GUV instrument and some preliminary results of the retrieved TCO. In particular, the TCO has been assessed by comparing the ratio of irradiances registered at 313 and 340 nm, with the corresponding ratio computed trough the Tropospheric Ultraviolet and Visible (TUV) radiation transfer model for different solar elevation angles and TCO. To avoid the effect of clouds, which are able to cause high-frequency variations in the measured solar irradiance and hence, impact the chosen ratio, the latter was approximated by a polynomial determined using trimmed regression before applying the comparison with the model. The results were compared with the TCO amounts provided by the Ozone monitoring instrument (OMI) on board the Aura satellite and the correlation coefficient between the OMI values and those retrieved by the GUV 2511 surface measurements, both referred to the Stara Zagora station, was found to be higher than 0.975.

**Keywords:** Ozone, ground-based measurements, validation

## Introduction

Routinely total column ozone (TCO) observations began with the design of the Dobson-spectrometer in 1924 (Dobson, 1968). After the International Geophysical Year 1957 global networks of ground-based ozone measurements were established, at first consisting only of Dobson spectrophotometers and later, after the design of the Brewer spectrophotometer (Kerr et al., 1985), instruments of this type were also included. Today the most important atmospheric monitoring network is the Global Atmosphere Watch (GAW) established by the World Meteorological Organization. One of the major contributors is the Network for the Detection of Atmospheric Composition Change (NDACC).

Dobson and Brewer instruments have a typical spectral resolution of 1 nm approximately. To perform Dobson spectrophotometer measurements the operator has to be very experienced. In contrast, Brewer instruments are working fully automated. The agreement of three calibrated by standard lamps reference Brewer instruments at Toronto, known as the Brewer triad, is better than 1% over a period of 20 years (Fioletov et al., 2005). Other Dobson and Brewer field spectrophotometers are calibrated during intercomparison campaigns. (e.g. Gao, 2001).

Since the 60ties low price pigmented glass filter instruments were developed (Gushin, 1963; Bojkov et al., 1994), where measured direct sun light and the ozone column were determined likely as for Dobson instruments. The design of highly sensitive detectors made it possible to obtain sky light spectrums during sunrise or sunset up to zenith angles of 94 deg. by ground-based SAOZ (Système d'Analyse par Observation Zénithale) UV-visible spectrometers (Pommereau and Goutail, 1988; Hendrick et al., 2011 and citations herein) based on the technique of Differential optical absorption spectrometry (DOAS) (Platt et al., 1979). Multi-filter rotating shadow band radiometers were designed for the visible spectral range (Harrison et al. 1994) and for the UV (Bigelow et al. 1998). Moreover rotating shadow band spectro-radiometers allow to provide diffuse and global spectral irradiance measurements at high spectral resolution (Harrison et al. 1999). By such measurements the total column of species as $NO_2$ and $H_2O$ can be retrieved in addition. Raptis et al. (2015) have proposed a method based on three dimensional Lookup tables of ratios of direct solar irradiances as function of the solar zenith angle, TOC and AOD to retrieve the TOC from rotating shadow band spectro-radiometers.

Here we will only note instrumentations as max-DOAS, ozone-Lidars and Balloon based measurements to retrieve vertical ozone distributions.

Dobson and Brewer spectrometers are widely used for the validation of other ground-based instruments (Dahlback, 2005) and for the validation of satellite ozone measurements (Roscoe et al., 1966; Richter et al., 1999; McPeters et al., 2007; Celarier et al., 2008). The error of the ozone retrieval by Brewer instruments is about 1% (Kerr et al., 1988). Balis et al. (2007) have obtained an averaged agreement better than 1% for OMI-TOMS data and better than 2% for OMI-DOAS data with ground-based observations. Stamnes et al. (1991) have shown that by the help of spectrometers with high resolution of about 0.5 nm the TCO and the cloud transmission can be determined very accurately. In the 90-ties broadband filter instruments were developed to increase the global coverage of the measurements. Broadband instruments with only a few filters allow also determining of the biological UV doses, the total ozone abundancies, and the cloud optical depths (Dahlback, 1996).

In February 2015 a Ground-based Ultraviolet Radiometer (GUV) 2511 was installed in Stara Zagora. The GUV 2511 instrument is designed for measurements of the downwelling global irradiances in six broadband channels and of the irradiance in the visible range from 400 to 700 nm. The instrument allows obtaining of the total column ozone (TCO) in the atmosphere, the determination of the UV-index and the retrieval of cloud optical thickness. In the paper the used methodology to derive TCO is described and the first results obtained by the GUV instrument are presented.

## Brief description of GUV 2511 instrument

The GUV instrument measures the global irradiances at 305, 313, 320, 340, 380, 395 nm with a full bandwidth of 10 nm at the half-maximum (FWHM) of the response function as well as the Photosynthetically Available Radiation (PAR) irradiance in the visible spectral range from 400 to 700 nm. The 313 nm filter was added in the GUV 2511 type instrument unlike the previous GUV 511 type instrument to ensure ozone determinations at greater zenith angles (of about 80°) where the absorption at 305 nm is very high (Booth, 1999). All channels have their own photodiode with amplifier. The instrument entrance window consists of a teflon diffusor on a quartz base. The cosine error of the instrument is smaller than 3% (7.5%) for zenith angles less than $65^0$ ($82^0$) (Bernard et al., 2005). A heater blanket placed in the instrument head stabilizes the photodiodes, filters and amplifiers at the temperature of $50^0$C. A portion of the heat is used to warm the diffuser and to keep it free from ice and snow. Melt water or rain on the diffuser and occluding ring is led outside by drain holes.

The instrument functioning is operated by a controller including the power management, the temperature control and the data transfer via the interface RS232. The instrument is connected with the controller by a 50 m long cable.

The main advantage of the GUV instrument series is that they have not moving components and therefore they work very stably. Moreover, the measurements are provided very fast. Our GUV-PC configuration achieved a measurement repetition frequency of 0.5 Hz. The instrument is operating in averaging mode with a 10 sec. averaging time.

The GUV instrument was installed in February 2015 on the roof of the Stara Zagora observatory. The observatory is located on a hill with an altitude of 430 m at 3 km to the North from the City Stara Zagora. The observatory is at about 250 m above the city and there are not other high buildings, thus a free view to the sky is ensured.

## Brief description of the OMI-Aura measurements

The Ozone Monitoring Instrument (OMI) together with other instruments is installed on the Earth observing system (EOS) Aura satellite platform. The Aura spacecraft sun synchronous orbit allows performing measurements over an Earth location approximately at the same mean solar time every day. OMI is an ultraviolet/visible (UV/VIS) nadir solar backscatter spectrometer with a spectral resolution of 0.42 nm in the UV-1 channel (270-310 nm) and 0.45 nm in the UV-2 channel (310-365 nm). The spectrometer provides nearly global coverage in one day with a spatial resolution of 13 km x 24 km (Levelt, 2006).

To retrieve TCO from the OMI measurements two different algorithms are used – one of them is based on the TOMS Version 8 algorithm developed for the ozone retrieval from the Total Ozone Mapping Spectrometer (TOMS) data (Bhartia and Wellemeyer, 2002). The second algorithm is based on the Differential Optical Absorption Spectroscopy (DOAS) technique. The satellite OMI ozone data were validated by the help of ground based Brewer and Dobson spectrometers. Important discrepancies depending from zenith angle were found between the OMI DOAS ozone and ground based data (Ballis et al., 2007; Buchard et al., 2008). In contrast Ialongo et al. (2008) haven't found discrepancies neither for the OMI-TOMS nor for the OMI-DOAS ozone data.

McPeters et al. (2007) have compared the OMI-ozone products with 76 Dobson and Brewer instruments located in the Northern Hemisphere. The authors have established that OMI-TOMS and OMI-DOAS was stable over the observation period of 2 two years with no drift in relation to the ground based ozone measurements and found OMI-TOMS (OMI-DOAS) averages 0.4% (1%) higher than the ground based ozone column average.

Here for the first look we used OMI-TOMS like ozone data to compare with the ozone results of our measurements. Gridded in steps of 1 degree by 1 degree OMI-TOMS like ozone data (OMTO3), for shortness called further OMI data, are available at https://ozoneaq.gsfc.nasa.gov/data/ozone/ (OMI-Aura Global Ozone Data). The gridded OMI data were bilinearly interpolated for the Stara Zagora observatory location. In the cases when the Stara Zagora location was close to the end of the OMI field of view swath the ozone map was interpolated for an additional location in close proximity to Stara Zagora allowing to use data from the next grid pixel. Normally when the locations were not at the end of the field of view the ozone differences at both locations were not greater than 2 DU. However, at the field end the differences were greater than 30 DU and the greater ozone value was selected. The Aura satellite passes over Bulgaria in about 13.30 o'clock local time with time differences of approximately 50 min. caused by the satellite orbit perturbation and deviations of the apparent solar time from the

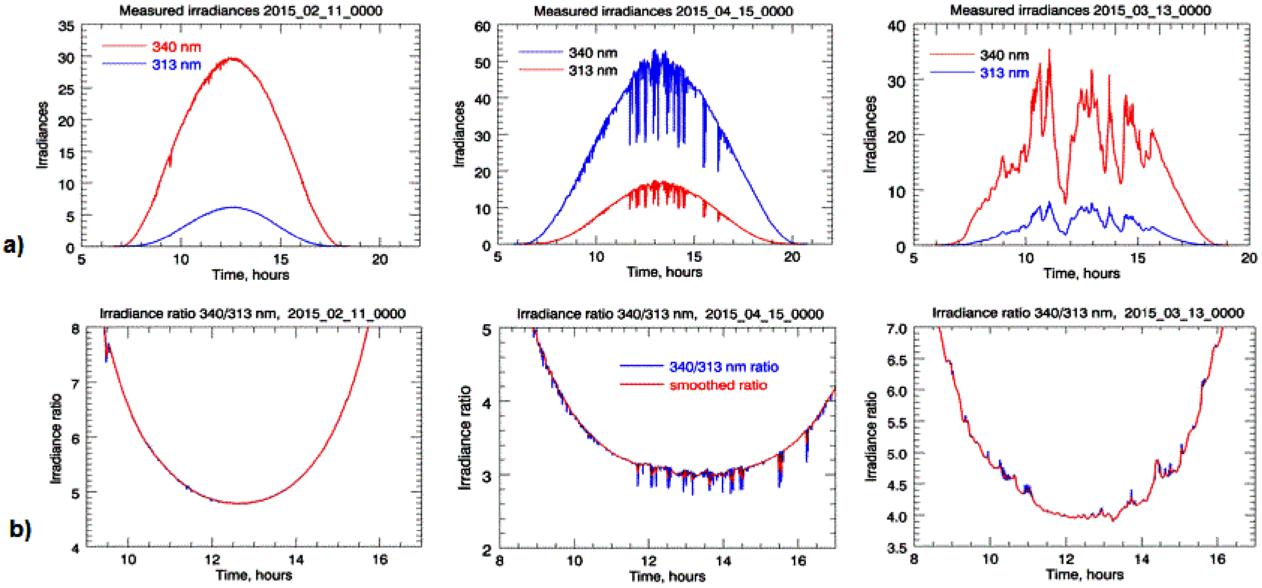

Fig.1. a) Measured irradiances at 313 nm and 340 nm, given in µmW/(cm2nm), for days with different cloudiness and b) the irradiances ratios (see text).

mean solar time. An additional time difference of approximately 40 min. results from the deviation of the view angle from the nadir direction.

Aura is part of the so-called A-train, consisting of five satellites with very close orbits, passing by a location in space for 8 min., which make it easy to use coincident additional information.

## Preliminary results

From February to the middle of June 2015 daily measurements with an integration time of 10 sec. were carried out with some interrupts for technical reasons. The irradiance maxima are observed at noon about 12:30 Local time or 13.30 Local summer time. The daily irradiances maxima change with the zenith angle. The minimum of zenith angle at the summer solstice is 19.25° and at the winter solstice the zenith angle not exceeded 66.12° at the latitude of Stara Zagora. The irradiances at clear days 3 hours before and 3 hours after noon are not smaller than 70% of the irradiance maximum.

Fig.1a shows the observed irradiances during days with different cloudiness: for an almost clear day (left), for a day with fast changing of the solar disc cloud cover (middle) and for a day with large cloud fraction (right). Fig.1b presents the irradiances ratios for the same days. The obtained irradiances are much more sensitive against changes of the overhead cloudiness in comparison with the observed irradiance ratios. High frequency variations are obtained for days with very fast changes of the cloudiness, probably connected with Cirrus clouds (in the middle of Fig. 1b). The irradiance variations registered at 340 nm are some more than about 40% of the maximum. For the 313 nm filter the variations achieve about one third of the maximum value at noon. And vice versa, the obtained irradiance ratio is much stable. The ratio varies only about 10%. In the case of strong cloudiness, the variations have a more low frequency character and the irradiance ratios decrease approximately by 0.5, which generates a great error in the TCO determination. Strong cloudiness often is connected to low clouds and raining.

## Methodology of TCO determination

### *Data pre-processing*

To reduce the cloudiness influence at first the average irradiances at 340 nm from 10:30 to 14:30 LT were calculated. If these radiances were lower than the estimated monthly value (approximately 1/3 of the average monthly mean for cloudless days) then we assumed, that the ozone determination would be wrong because of strong cloudiness. In the second stage the ratios were smoothed by a running boxcar over 17 values, e. g. over 170 sec. Under strong cloudiness the irradiance ratios variations decrease only up to 0.5 (see e.g. the right panel of Fig.1b). Therefore an upper envelope for each daily ratio development was determined. A detailed description of this procedure was given by Werner et al. (2015).

The TCO retrievals were successfully provided for 379 days during the studied time period. Strong cloudiness was established for 54 days. For six days the polynomial shows strong oscillations, which does not allow TCO determination with high accuracy. The number of almost cloudless days was obtained by inspection of the daily progress of the 340 nm irradiation. By this subjective evaluation 103 days were determined as almost cloudless. For 249 days (81 of them are cloudless days) coincidences with OMI measurements were found.

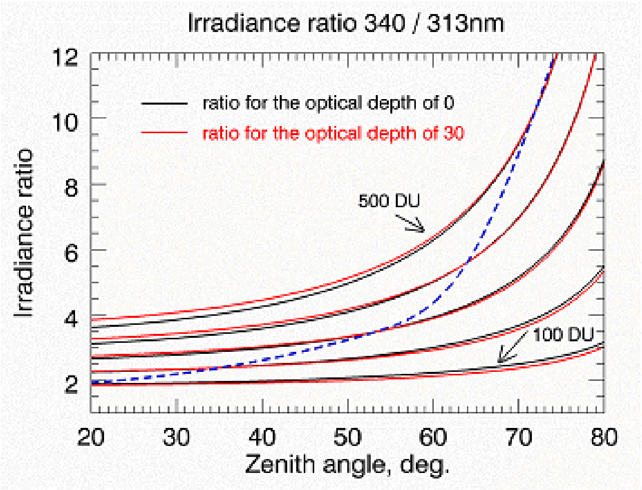

Fig.2. Calculated Stamnes table (ground albedo of 0.2) for two optical depths for the Stara Zagora location. The irradiance ratios are shown for TCO from 100 to 500 DU in steps of 100 DU and for zenith angles from 20° to 80°.

### *Calculation of Stamnes table for Stara Zagora location*

GUV instruments measure the solar global horizontal irradiance consisting of the diffuse and the direct component of the solar irradiance. The ratio of the irradiances at different wavelengths depends on the scattering properties of the air particles and molecules and also on the solar elevation, e.g. on the absorbing path length through the atmosphere. Therefor TCO cannot be determined directly by the observed irradiance ratios based on the Beer-Lambert law as it is possible for Dobson and Brewer spectrophotometers for direct solar measurements. To determine TCO from UVA/UVB ratios simulations by radiation transfer models including the UV spectral range are necessary. Here we used Tropospheric Ultraviolet and Visible (TUV) model, version 4.1., developed by Madronich (1993), where the Rayleigh scattering parameters were calculated through an improved algorithm for standard atmospheric conditions (Tomasi et al. ,2005; Petkov et al., 2006). The spectra were calculated for the Stara Zagora location for different TCO from 0 up to 700 DU with a step of 20 DU and zenith angles from 20° up to 90° with a step of 1° . A ground albedo of 0.2 was used as input parameter. We used the climatological mean ozone profile of the U.S. Standard Atmosphere 1976, since the real ozone profile has a noticeable influence on the ozone retrieval only for large zenith angles (Høiska et al., 1997). The mean ozone profile is part of the TUV model library. The obtained spectra were multiplied with the relative filter response functions, approximated by a Gaussian with 10 nm FWHM. The irradiance ratios for wavelengths 340 nm and 313 nm were calculated and the ozone content values were determined by interpolation of the calculated with the cloud optical depth $\tau=0$ Stamnes table for the zenith angles corresponding to time moments, when the measurements were performed. The zenith angles were calculated by an astronomical algorithm (see Meeus, 1993). Stamnes tables also called Look up tables (LUT).

For the TCO retrieval by interpolation of the Stamnes table for the corresponding zenith angles at 13.30 LT the ratios of the envelopes are used. The irradiance ratios depend on the real filter functions (more correct from the spectral instrument responses).

As pointed out above the TCO determination was provided for cloudiness days only if the irradiation intensity at the wavelength 340 nm was greater than 0.3 of the one at cloudless days, corresponding to an optical depth smaller than $\tau=30$. The resulting Stamnes table for $\tau=30$ is shown in Fig. 2 (dashed red line) together with the one for $\tau=0$ (black line). The cloud effect on ozone retrieval depends on the ozone level itself and on the zenith angle of the observations for a given irradiation ratio. The blue line in Fig. 2 separates regions with overestimation of TCO (above the line) and with underestimation (below the line) of TCO. The resulting overestimation of ozone is about 10 DU for high Sun and an ozone level of 300 DU and smaller than 5 DU for low Sun and lower ozone levels matching to the found by Stamnes et al. (1991) cloud effect.

As mentioned above our GUV instrument is not validated against a standard normal (Dobson or Brewer) spectrometer. Therefore we regressed our TCO results with the OMI satellite ones using different wavelength maxima for the 313 nm filter in the Stamnes table calculation procedure. The best TCO results were obtained with filter centered at 340 nm and at 313.55 nm. This approach does not allow us to specify a systematic error between the two measurements – GUV and OMI. A sensitivity of the results to different albedo between 0.05 and 0.5 was not found.

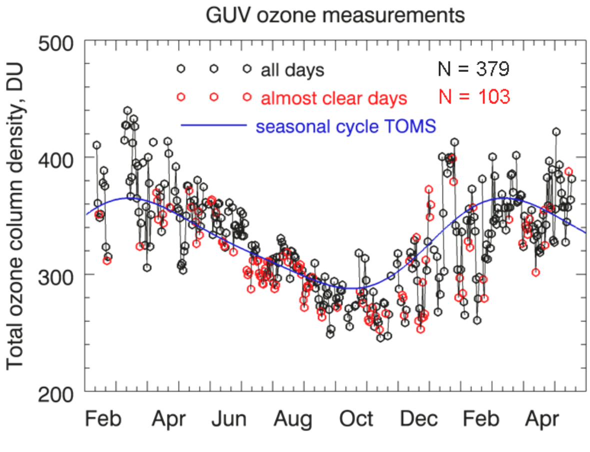

Fig.3. TCO for Stara Zagora determined for the observation period since February 2015 (circles connected by a continuous line for uninterrupted measurements). The TOMS TCO seasonal means are presented by a thick blue line. The TCO values for Stara Zagora at clear days are shown by red circles.

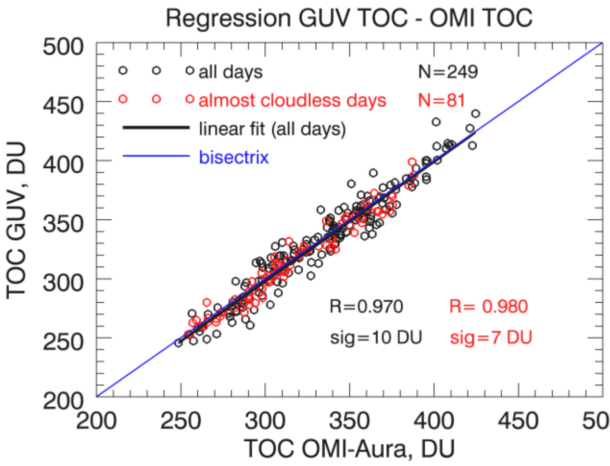

Fig.4. Linear relation between the TCO GUV and the TCO OMI data. The thick black line presents the regression fit.

## Results

The determined TCO time series during the period since the GUV 2511 installation in February 2015 up to the end of May 2016 is shown in Fig.3. Consecutive daily GUV ozone values are connected by a continuous thin black line. Long Interruptions mark periods without data for technical reasons. Interruptions of some days could be a result of bad weather conditions with strong cloudiness. The observed TCO (Fig. 3) follows closely the multi-annual seasonal mean represented by a thick continuous blue line, determined from TOMS Earth Probe measurements from 1996 to 2005. An abrupt ozone maximum in spring and a decrease to the minimum in autumn ([see e.g. Mendeva et al. 2010) and higher variability during spring can be seen as it is typical for midlatitudes.

Fig. 4 shows both our ground based TCO and OMI satellite ozone data for days when measurements with both instruments are available. The figure represents the result of the linear regression of our estimated GUV 2511 TCO data against the OMI data. It is clearly seen that our found ground-based TCO values reproduce the satellite OMI TCO very well. The slope of the best fit line between GUV and OMI TCO for all days is 1.013 with a 1σ error of 0.031. At the significance level of 0.05 the slope is not distinguishable from 1. The found correlation coefficient is 0.97, meaning that the explained variance of the deviations of the GUV ozone values from the OMI ones is about 0.94. This result is comparable to the found correlation between the Brewer instrument TCO located at the Rome station and the OMI-TOMS TCO values of 0.97 for 574 coincident observations reported by Ialongo et al., (2008). The observed TCO at almost cloudless days, shown by red dots, are better correlated to the OMI TCO. The explained variance is 0.960. While the agreement between the GUV and OMI TCO is very good, it is evident that a cloud influence in our retrieval procedure remains. Bogeat et al. (2012) were reported overestimations of GUV ozone measurements at cloudy days during the comparison campaign in April 2010. Dahlback et al. (2005) have observed underestimations of TCO at the cloudy day 197 of 2002 in Oslo.

The relative errors of our TCO determinations (related to the OMI TCO) are presented in Fig. 5. The maximal errors do not exceed 10%. The mean standard deviation (1-σ) is about 3.1%. This means that 95% of the errors of the daily TCO are smaller than 6.2%. The mean error is not ideally zero, which is not a systematic error. The reason is the not perfectly chosen wavelength alinement of the filter 313 nm maximum.

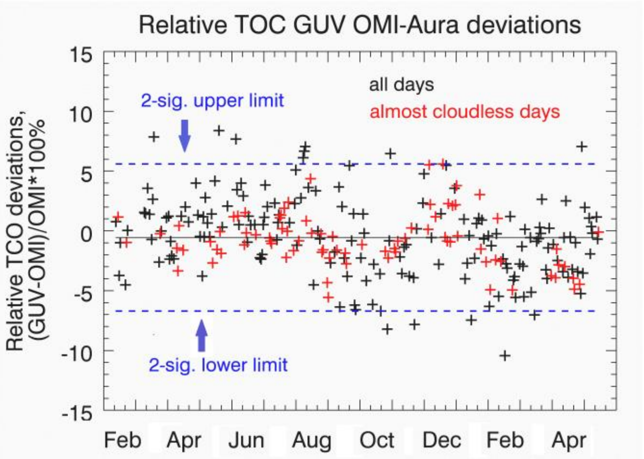

Fig.5. Obtained relative deviations of TCO by the GUV instrument from the OMI data. The horizontal continuous black line presents the mean error, which is expected to be close to zero. The dashed blue lines mark the 2-σ confidence interval.

A similar standard deviation of 3.5% of daily averaged ozone measurements performed with a NILU-UV filter instrument in comparison with Brewer ozone measurements was reported by Kazantzidis et al. (2009) for the Athena and Thessaloniki stations.

As it is seen in Fig. 5, all relative deviations between the GUV and the OMI-TOMS TOC for almost cloudless days lie within or exactly at the 2-σ limits calculated using all TOC values for all days. At almost cloudless days the mean 1-σ deviation is only 7 DU corresponding to a relative error of approximately 2.2 %. Most of TCO retrievals with an error greater than about 8% were obtained at days with very strong cloudiness. Masserot at al. (2002) have stated that for a few cases the differences of TOMS satellite TCO and ground-based TCO can reach 10 to 15%. Dahlback (1996) found by computer simulation an overestimation of 20 DU for a high albedo of 0.8 and a cloud layer between 2 and 4 km with an optical depth of τ=100 if the TCO is determined by the ratio of the irradiances at the two wavelengths 320 nm and 305 nm if the cloudiness is not taken in consideration. For an albedo 0.0 and for τ=100 the estimated error is less than 2 DU. Dahlback et al. (2005) reported an underestimation of the TCO of about 5 DU obtained by the NILU-UV filter instrument in comparison with the Brewer in Oslo at the day 197 of the year 2002 when the Cloud transmission factor (CLF) was about 50 %. (The CLF is defined as the ratio of the measured irradiance and the calculated irradiance for a UV-A channel for clear sky with no aerosols and zero albedo, both for the corresponding to the measurements zenith angle.) In the early evening when the CLF was 70% the difference reached approximately 8 DU.

Meyer et al. (1998) have shown that the use of LUT for τ=0 leads to overestimation of the TCO in case of optically and geometrically thick cloud during a thunderstorm, and to underestimation of TCO resulting from redirection of photons scattered within the cloud and emerging downward from its base. For more detailed discussion also for effects as aerosol properties, absorption coefficients and stratospheric temperature (see Slusser et al., 1999).

Clouds can cause different TCO errors (overestimation and underestimation). A more careful consideration of the cloud influence on TCO and implication in our TCO retrieval algorithm is needed.

## Conclusions

The retrieved TCO data from ground based UV measurements by help of the GUV 2511 instrument at Stara Zagora are in good accordance with satellite OMI-Aura data obtained by the TOMS v.8 algorithm. Between our estimated ground-based TCO and the satellite OMI TCO data a correlation of 0.975 is found. However a systematic deviation of both instrument results cannot be established because our instrument is not calibrated against a Dobson spectrometer or Brewer instrument. Regardless our UV irradiance measurements allow the determination of daily TCO with an error of 3.1%, corresponding to about 10 DU. It was found a cloud influence on the TCO. An improvement of the retrieval procedure taking into account an estimation of the optical depth is envisaged. A detailed analysis in order to obtain more accurate filter functions is necessary to improve the quality of the TCO data. Also other filter combinations have to be tested to retrieve TCO. The instrument has to be calibrated by the help of standard ozone spectrometers. We state, that high quality ground-based ozone time series are to be expected for the Stara Zagora location in the future.

## Acknowledgement

The GUV instrument was provided by the project BG161PO003-1.2.04-0053 "Information Complex for Aerospace Monitoring of the Environment" (ICASME) implemented with the financial support of Operational Programme „Development of the Competitiveness of the Bulgarian Economy 2007-2013", co-financed by the European Regional Development Fund and the national budget of the Republic of Bulgaria.

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