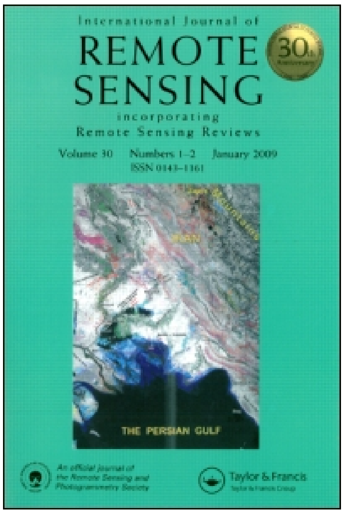

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Evaluation of ozone column measurements over Greece with NILU-UV multi-channel radiometers

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Measurements of total ozone over Greece have been performed with NILU-UV multi-channel radiometers since 2005. In this study, the measurements at two stations, Thessaloniki and Athens, of the ultraviolet (*UV*) monitoring network are compared with ozone values derived from Brewer spectroradiometers at the same sites. The overall percentage difference on daily averages (including measurements for solar zenith angle below 70°) is below (1 ± 3.5)% at both sites. The cloud modification factor (CMF), specified as the ratio of measured irradiance with a model-derived cloud-free value, is being used to examine the effect of cloudiness on the retrieved ozone abundances. During days with average CMF values above 0.7, the difference in total ozone measurements is within ± 5%. The use of momentary ozone values from the radiometers with this limitation in CMF reduces the difference of the average values to (0.0 ± 2.2)% for Athens and (0.21 ± 2.8)% for Thessaloniki.

1. Introduction

In recent years, multi-channel radiometers have been used in national or international networks, or at specific sites, since they are able to provide biologically effective doses for a variety of action spectra, photolysis rates, total column ozone amount and cloud attenuation at high temporal resolution (Bigelow *et al*. 1998, Di Meno *et al*. 2002, Johnsen *et al*. 2002, Seroji *et al*. 2004, Bernhard *et al*. 2005, Lakkala *et al*. 2005, Lovengreen *et al*. 2005, Bhattarai *et al*. 2007, Cortesi *et al*. 2007, Kazadzis *et al*. 2007, Kazantzidis *et al*. 2007, Myhre *et al*. 2007). In parallel, new methodologies have been proposed to reduce the measurement uncertainties, and international campaigns were organized to improve radiometer calibration standards (Dahlback 1996, Bernhard *et al*. 2005, Diaz *et al*. 2005, Johnsen *et al*. 2008).

The determination of the total ozone abundance and the effective cloud transmission from global ultraviolet (UV) spectral irradiance measurements was proposed by Stamnes *et al*. (1991). They used two wavelengths, one with appreciable absorption by ozone (305 nm) compared with the other one (340 nm), and they compared their model-derived ozone columnar values with *in situ* measurements in Antarctica. Dahlback (1996) proposed a new method, based on a four-channel radiometer, to

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derive ozone, cloud transmission and UV dose rates. The relative difference from total ozone 1 year measurements by a Brewer and a Dobson instrument in Oslo, Norway was (0.3 ± 2.9)%, while the standard deviation was reduced by 1% when ozone measurements under clear sky and solar zenith angles below 60° were included. More recently, the comparison of 3 year measurements at the same site revealed percentage differences less than 2% (Dahlback *et al.* 2005). Additionally, the influences of clouds and ozone vertical profiles on ozone measurements from the radiometer were found to be small for cloud transmittances down to 30% and solar zenith angles below 65°. Bernhard *et al.* (2005), using a method similar to that described by Stamnes *et al.* (1991), compared ozone values from the multi-channel radiometers and the spectroradiometers of the US National Science Foundation UV monitoring network and reported percentage differences of the same magnitude (2 ± 2)% for solar zenith angles below 80°.

In this study, after the first 3 years of operation, the accuracy of the total ozone estimates by the NILU-UV multi-channel radiometers over Greece is examined. Comparisons between daily averages from Brewer spectroradiometers and radiometers at the first two stations of the Greek UV network are presented, and the impact of cloudiness is discussed.

2. Measurements

The Greek UV network (www.uvnet.gr) was designed to geographically cover Greece and Cyprus, comprising eight satellite stations, distributed at locations with different environments, and a central station located at Thessaloniki, where a suite of spectral and broadband radiation and other related measurements are performed. The network provides online 1 minute averages (with standard deviation) of channel irradiance from all stations through a newly designed algorithm. A series of products, such as total ozone, cloud cover assessment and several radiative quantities (Commission Internationale de l'Eclairage (CIE)-weighted UV dose rates, integrated UV-A, UV-B and visible irradiances) are being provided. More details about the network can be found in Kazantzidis *et al*. (2007).

The NILU-UV multi-channel radiometers provide UV irradiance measurements at five wavelength bands centred at 302, 312, 320, 340 and 380 nm, with full width at half maximum (FWHM) of approximately 10 nm. In addition, a sixth channel measures photosynthetic active radiation (PAR) between 400 and 700 nm. The technical details of the instrument are described in detail in Hoiskar *et al*. (2003).

All instrument channels were characterized as to their spectral response with a powerful 1000 W xenon lamp and a double monochromator. The deconvolution procedure, as suggested by Bernhard *et al.* (2005), was followed for the conversion of the measured signals to spectral irradiance at the central wavelength of each channel. The absolute calibration factors were determined by comparison to the spectral irradiance measurements from a double monochromator Brewer spectroradiometer #086, operating at Thessaloniki. The procedures for the absolute calibration of the instrument and the cosine correction methodology are presented in Garane *et al.* (2006). According to Bernhard *et al.* (2005), the agreement of NILU-UV derived irradiance with spectral measurements is within ± 5% for solar zenith angles smaller than 85°.

The NILU-UV #04103 and #04105 instruments have been established since December 2004 at Thessaloniki (Laboratory of Atmospheric Physics, Aristotle University of Thessaloniki, 40.6° N, 23° E) and Athens (Biomedical Research

Foundation of the Academy of Athens, 38° N, 23.8° E), respectively. The NILU-UV #04103 radiometer is used routinely to perform measurements next to the reference spectroradiometer, while #04105 radiometer returns every 6 months for calibration. The NILU-UV calculated ozone values presented in this study are derived from UV irradiance measurements at 305 and 340nm. Based on the methodology suggested by Stamnes *et al*. (1991), model-calculated look-up tables of global irradiance were used for the estimation of total ozone. The cloud-free irradiance was calculated from the UVSPEC model, using average values for aerosol conditions at Thessaloniki (Mayer and Kylling 2005). The average aerosol optical depth at 340 nm and the Angstrom exponent were assumed to be equal to 0.4 and 1.4, respectively (Kazadzis *et al*. 2007). Spectrally independent values, 0.9 and 0.7, were used for the single scattering albedo and the asymmetry factor of aerosols, respectively (Bais *et al*. 2005). Similar aerosol conditions were revealed from measurements in the UV region at Athens (Jakovides *et al*. 2005). For all calculations, mid-latitude standard vertical profiles for air density, ozone and temperature were used (USSA 1976) and the surface albedo was set to 0.05, independent of wavelength. The overall uncertainty of the method is estimated to be less than ± 5% for cloud-free sky conditions and solar zenith angles below 70° (Stamnes *et al*. 1991, Bernhard *et al*. 2003).

For higher solar zenith angles, the impact of cloudiness, the vertical profile of ozone and temperature, the imperfect cosine response and the absolute calibration error of the instrument significantly reduce the accuracy of the method results. The measured ozone at Thessaloniki in the lower troposphere (0–5 km) is, in most cases, higher than standard profiles (e.g. Kourtidis *et al*. 2002, Galani *et al*. 2003, Varotsos 2005). The corresponding effect in the calculation of UV irradiance at 305 nm reaches 5% and 7% for 30° and 70°, respectively (Kazantzidis *et al*. 2005). According to laboratory measurements, the uncertainty in measurements introduced from the NILU-UV imperfect cosine response is diminished for solar zenith angles below 70° (Kazantzidis *et al*. 2006). For these reasons, only total ozone measurements below 70° are compared in this study. This limitation does not affect the measurement availability, since the solar zenith angle at local noon for both sites is lower than 63° throughout the year.

Due to the multiple impacts of global ozone dynamics on environmental safety (e.g. Kontratyev and Varotsos 1996), measurements of total ozone and ultraviolet irradiance have been routinely performed at the same sites or in the surrounding region since the 1980s (Bais *et al*. 1993, Varotsos 1994, Zerefos *et al*. 1997, Efstathiou *et al*. 1998). In this study, we used total ozone measurements performed at the Laboratory of Atmospheric Physics of the Aristotle University of Thessaloniki, with Brewer #005 single spectroradiometer, which has been in operation in Thessaloniki since 1982 (e.g. Bais *et al*. 1996, Zerefos 2002). Since 2003, Brewer #001 monochromator measures the columnar amount of ozone in Athens. The spectroradiometer is calibrated regularly by means of a standard radiometer of the same type. The last two calibrations were performed in July 2004 and June 2007 by the travelling standard Brewer #017 (Zerefos and Eleftheratos 2007).

3. Results

3.1 Daily ozone measurements

The percentage variability of the difference between the daily averages of total ozone, measured from NILU-UV radiometers and Brewer spectroradiometers, is presented

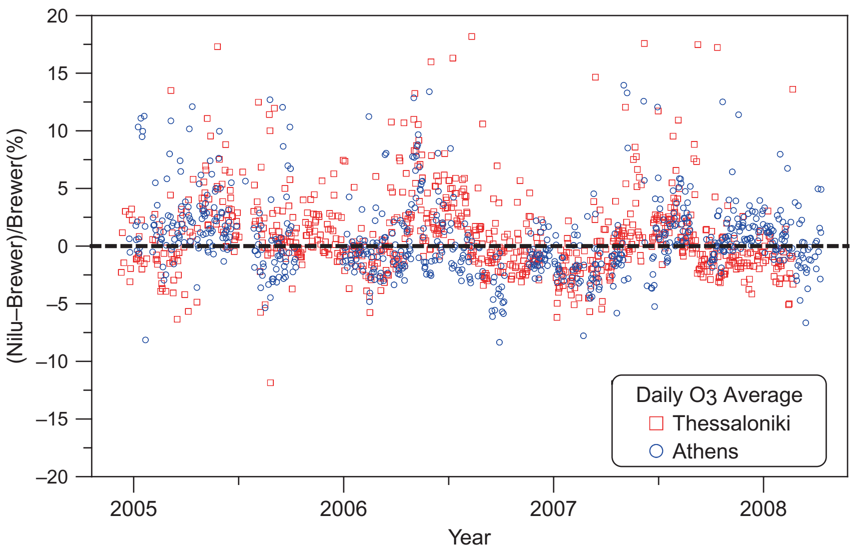

Figure 1. Percentage difference of the daily averages of total ozone, as derived from NILU-UV and Brewer measurements at Thessaloniki and Athens, for the 2005 to 2007 time period.

in figure 1. For all instruments, the available momentary ozone measurements at solar zenith angles below 70° were included in the calculation of the daily average.

At both sites, the total ozone is scarcely overestimated by the radiometers; the average difference is less than 1% (0.90% at Thessaloniki and 0.76% at Athens) and the standard deviation is around 3.5%. The results are comparable with similar studies at other sites, reporting differences within ± 2% (Bernhard *et al.* 2005, Dahlback *et al.* 2005). Increased percentage differences could be mainly attributed to the effects of clouds on NILU-UV global irradiance measurements, and will be further discussed in the next paragraph. The differences are generally higher during springtime, by almost 3%, when compared with autumn values. This pattern is similar with the total ozone annual behaviour (more in spring and less in autumn). It could probably be attributed to the uncertainty of the method to accurately calculate the global irradiance at 305nm from the corresponding NILU-UV channel; slight uncertainties in the measured spectral response could introduce such errors because of the dissimilar ozone absorption of irradiance in the UV-B wavelengths. The penetration of moisture into the detector followed by slow drying could be one more reason (Lakkala *et al.* 2003).

3.2 Modelled estimated effect of cloudiness

The effect of cloudiness on the ozone estimation from the NILU-UV radiometers was estimated by the calculation of the cloud modification factor (CMF). This factor is described as the ratio of irradiance under cloudy conditions with the corresponding cloud-free value, considering that all other atmospheric conditions remain the same. Model-calculated values of irradiance at 340 nm are compared with NILU-UV values at the same wavelength in order to estimate the cloud effect on ozone calculations. At this wavelength, the ozone absorption is diminished, but the impact of clouds is not spectrally independent in the UV region, even under overcast conditions (Mayer *et al.* 1998, Schwander *et al.* 2002, Lindfors and Arola 2008). In addition, the effect of aerosol optical depth (when it is different from its average value) was also included in the CMF calculation. Therefore, the presented CMF should be only considered as an indicator of the cloud effect in estimation of total ozone.

CMF values close to 0 are representative of radiative conditions under heavy cloudiness, while the values are expected to be close to 1 for cloud-free and average aerosol conditions. In some cases, CMF values higher than 1 correspond to cases with low aerosol conditions or with enhanced irradiance due to scattered cloudiness (Sabburg and Long 2004).

The percentage differences in daily averages of total ozone from Brewer and NILU-UV measurements is presented as a function of the average value of CMF in figure 2. At both sites, CMF values up to 1.3 are calculated, indicating the high variability of aerosol optical depth and cloudiness. For the great majority of days with average CMF values above 0.7, the difference in total ozone measurements is within $\pm$ 5%. For lower CMF values, the NILU-UV measurements are sensitive to clouds and the measurement error is increased. At both sites, the days with average CMF value below 0.7 represent 15% of the total number of measuring days ($\sim$55 out of 365 days of the year). Since the two stations, situated in the eastern Mediterranean, are not affected dominantly by persistent heavy cloudiness, the momentary NILU-UV ozone values were also examined. Using the same limitation for CMF ($>$0.7) and solar zenith angle ($<$70°), there was only 1 day and 4 days per year at Athens and Thessaloniki, respectively, with less than 30 accepted momentary ozone values. In this case, the average percentage difference between NILU-UV and Brewer daily mean total ozone is reduced to (0.0 $\pm$ 2.2)% for Athens and (0.21 $\pm$ 2.8)% for Thessaloniki. The proposed limitation for CMF is significantly higher than the proposed values by Dahlback *et al.* (2005). In contrast, the CMF range is about the same because the effect of aerosols and clouds is different on model calculations and measurements.

4. Conclusions

Two decades after the implementation of the Montreal Protocol, stratospheric ozone loss is still severe over the poles, although the observations suggest that the concentrations of the ozone depleting substances in the atmosphere have started to decrease. Additionally, it is crucial to understand the interconnections between climate change and ozone depletion, since the projections of future ozone abundances are sensitive to changes of climate (WMO 2007).

Remote sensing played a key role in identifying and explaining the ozone depletion over the Antarctic (Farman *et al.* 1985). During the last few decades, ground-based and satellite measurements form the basis to establish an ozone climatology or trends on a local and global scale, respectively (e.g. Chandra and Varotsos 1995, Paul *et al.* 1998, Stahelin *et al.* 1998). The success of the Montreal Protocol is largely based on the scientific progress made by the use of all those measurements. In recent years, the increased need of satellite validation and investigation of ozone variability with higher spatial and temporal resolution, have led to the establishment of ozone monitoring networks equipped with multi-channel radiometers. As a result, specific methodologies have been applied to ensure that the accuracy of these instruments could be confirmed against ground-based measurements of known quality.

In this study, total ozone values, measured for 3 years (2005–2007) by NILU-UV radiometers of the Greek UV network at Thessaloniki and Athens, are compared with Brewer measurements for solar zenith angles below 70°. The percentage difference of

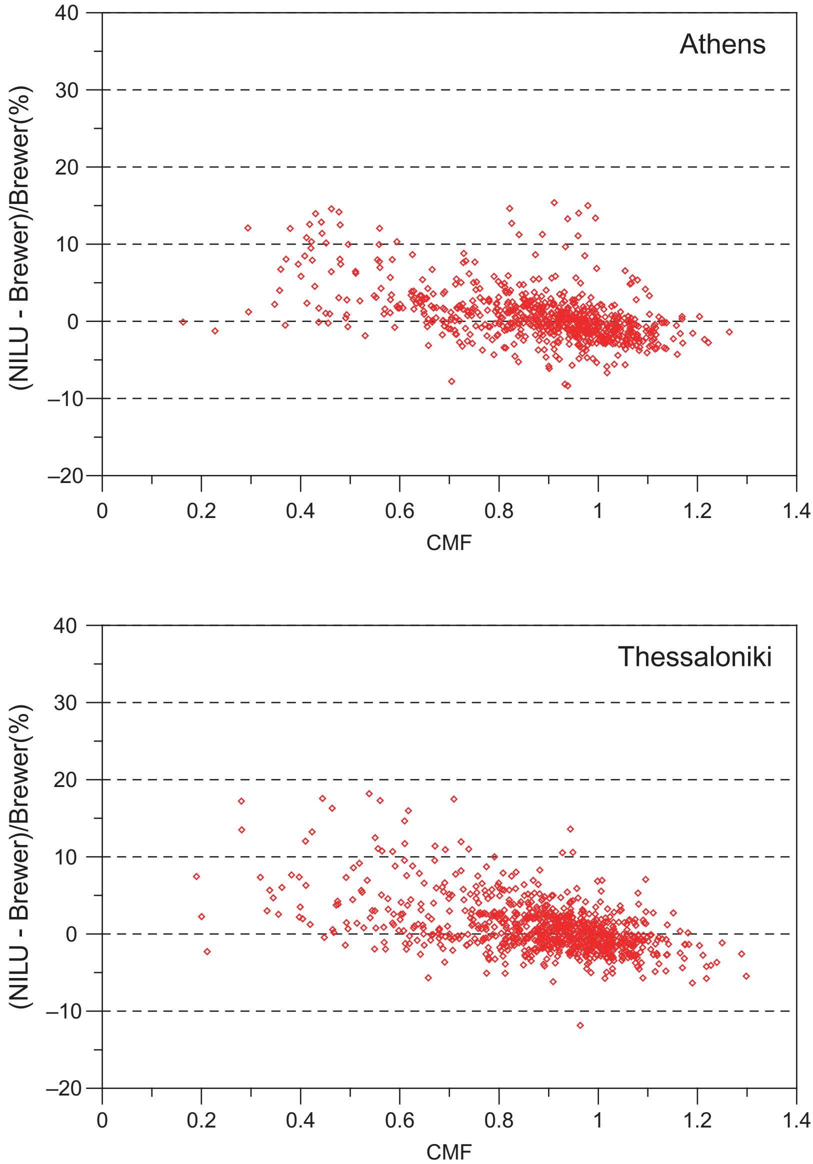

Figure 2. Percentage difference between NILU-UV and Brewer ozone columnar values as a function of the measured cloud modification factor (CMF) from NILU-UV measurements of global irradiance at 340 nm.

the daily averages is (1 ± 3.5)%. Additionally, an annual variation of the difference by 3% is revealed, which is probably related to the corresponding ozone variability throughout the year.

The ratio between measured and modelled global irradiance at 340 nm, the CMF, is calculated and used to examine the effect of cloudiness on the total ozone measurements from the radiometers. For days with average CMF values above 0.7, corresponding to 85% of the total days with measurements, the above-mentioned percentage difference is within 5%. The use of the same limitations for CMF (>0.7) and solar zenith angle (<70°) on the momentary ozone values, reveals that only 1 to 4 days of the year are excluded from the analysis. Additionally, the average percentage difference between NILU-UV and Brewer daily mean values of total ozone is reduced to (0.0 $\pm$ 2.2)% for Athens and (0.21 $\pm$ 2.8)% for Thessaloniki.

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