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An improved algorithm for the determination of aerosol optical depth in the ultraviolet spectral range from Brewer spectrophotometer observations

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Abstract
Methods to derive aerosol optical depth in the UV spectral range from ground-based remote-sensing stations equipped with Brewer spectrophotometers have been recently developed. In this study a modified Langley plot method has been implemented to retrieve aerosol optical depth from direct sun Brewer measurements. The method uses measurements over an extended range of atmospheric airmasses obtained with two different neutral density filters, and accounts for short-term variations of total ozone, derived from the same direct sun observations. The improved algorithm has been applied to data collected with a Brewer mark IV, operational in Rome, Italy, and with a Brewer mark III, operational in Lampedusa, Italy, in the Mediterranean. The efficiency of the improved algorithm has been tested comparing the number of determinations of the extraterrestrial constant against those obtained with a standard Langley plot procedure. The improved method produces a larger number of reliable Langley plots, allowing for a better statistical characterization of the extraterrestrial constant and a better study of its temporal variability. The values of aerosol optical depth calculated in Rome and Lampedusa compare well with simultaneous determinations in the 416–440 nm interval derived from MFRSR and CIMEL measurements.

Keywords: aerosol optical depth, ultraviolet radiation, Brewer spectrophotometer

1. Introduction

Aerosols play an important role in the Earth radiative budget. They can affect climate directly, through scattering and absorption of the incoming solar radiation and outgoing terrestrial radiation, and indirectly, acting as cloud condensation nuclei. The second effect is connected to the potentiality of aerosol in interacting with cloud formation, lifetime, and optical properties. Owing to the large variability in aerosol composition, shape, microphysical and optical characteristics and distribution, the global effect on climate is one of the largest unknown issues of the Earth's climate system [1]. For example, the direct radiative forcing at the top of the atmosphere in an area characterized by complex aerosol transport phenomena, like the Mediterranean sea, can be positive, null or negative, depending on the aerosol properties [2].

Aerosol optical properties are relatively well known in the visible spectral range. Little information is available on

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the optical properties of atmospheric aerosol in the ultraviolet spectral region. Some studies have discussed the impact of aerosol on the UV transfer in the atmosphere, e.g. [3–5]. Recently, it has been observed that desert dust and aerosols from biomass burning can significantly reduce surface UV levels [6]; moreover, in highly polluted areas, absorption of solar UV radiation by urban anthropogenic aerosols produces a reduction of surface UV irradiance; this reduction may mask the surface UV increase associated with low total ozone episodes [3].

Radiation measurements in the UVB spectral interval 280–320 nm are difficult because of the strong ozone absorption, and consequent large signal dynamical range. Dedicated instruments, such as the Brewer spectrophotometer, have been designed to perform measurements of UV radiances and irradiances. The existing network of Brewer spectrophotometers (about 140 instruments distributed worldwide mainly dedicated to total ozone monitoring) could contribute to increasing the spatial coverage of aerosol observations. These advantages have stimulated the development of algorithms to retrieve aerosol optical depths (AODs) from Brewer observations of direct sun radiances. Analyses of decade-long Brewer AODs have been presented, e.g. [8, 9, 11]. Two different approaches for the aerosol optical depth derivation from direct sun (DS) measurements have been proposed: the first one is based on the absolute calibration of direct sun observations, e.g. [10, 11], and the second one is based on the experimental determination of the extraterrestrial radiance by the Langley plot (LP) method e.g. [8, 9, 12–14].

In this paper we describe an improvement of the conventional LP method. Starting from the algorithm described in [14], we implemented a more accurate calibration scheme by performing LPs using data obtained with two different neutral density filters (NDFs) instead of one (thus expanding the interval of atmospheric air masses used in the LP), and by removing the effects of short-term ozone variability on LPs by subtracting from each measured radiance the ozone absorption. The new methodology was applied to Brewer observations carried out at the remote-sensing ground-based stations of Rome and Lampedusa. The retrieved AODs at 320.1 nm have been compared with measurements of a MultiFilter Rotating Shadowband Radiometer (MFRSR) [15] and a CIMEL sun photometer, operating in the frame of the Aerosol Robotic Network (AERONET) [16] at the same sites.

2. Instruments and measurement procedures

The Brewer spectrophotometer has been designed to perform measurements of total ozone, total sulfur dioxide, total nitrogen dioxide, spectral global irradiance and ozone vertical profiles by the Umkehr method.

A Brewer system is composed by the Brewer spectrophotometer, an azimuth tracker, and a controlling PC equipped with dedicated software. The system can be programmed for continuous operation by means of a *schedule*, in which observations are started depending on the solar zenith angle. The Brewer spectrophotometer performs measurements of global irradiance, zenith radiance, and direct sun radiance. In the normal use of the instrument, DS measurements are dedicated to the determination of total ozone.

Different models of Brewer exist, depending on instrument evolution and features. For a detailed description of the instrument, see [17].

In this study we use data collected with Brewers mark III and mark IV. The main difference between the two models resides in the monochromator optical design: mark IV uses a single monochromator with a 1200 lines mm^{-1} diffraction grating, while mark III uses a double monochromator with two 3600 lines mm^{-1} diffraction gratings. The mark-III stray light rejection is higher than for mark IV [18]. The mark-IV stray light rejection is achieved by means of a nickel sulfate filter that produces a significant temperature dependence of the measured signals [9, 19, 20]. The effect of stray light on AOD retrievals has been investigated e.g. in [22] and [23]. Arola and Koskela [23] show that at 306.5 nm, for an airmass range of 3–5, about 1–2% of the signal is due to stray light. The stray light influences the determination of the extraterrestrial constant, and the derived AOD. The influence of stray light on Brewer measurements above 313.5 nm, and for double monochromator Brewers, is negligible.

For each DS procedure, direct solar radiances are measured at six wavelengths (303.2, 306.3, 310.1, 313.5, 316.8 and 320.1 nm). The first wavelength is used for spectral calibrations, while combinations of the natural logarithm of the radiances at the other five wavelengths are used to compute total ozone. Depending on the incoming light intensity, the instrument automatically selects one among an open aperture and five different neutral density filters to maintain the photomultiplier within its linear response interval. The use of neutral density filters of different attenuations does not affect the total ozone measurement, which is calculated from ratios between logarithms of radiances. This aspect is of great importance for AOD retrievals.

The direct sun measurements at the five operational wavelengths are used in the determination of the aerosol optical depth. The method used for the calibration of the Brewer and the operational algorithm for the optical depth retrieval are described in the next section. The application of the algorithm to data from two Brewers is described in section 4.

3. Algorithm and calibration

The adopted method, as previously mentioned, is based on the Langley plot. The Langley plot is the linear regression of the logarithm of the measured radiance versus the airmass. The extraterrestrial instrumental constant is determined from the plot as the extrapolated radiance at zero airmass. In the Langley method the reference radiation source is the sun itself, and a relative radiometric calibration, i.e. in instrumental units, is obtained.

The main limitation of the method is that stable atmospheric conditions (i.e. constant aerosol optical depth, and, in the UV, total ozone and atmospheric pressure) are needed throughout the selected half day to obtain reliable extraterrestrial constants. As discussed in [11] and [24], short term ozone variations, in particular, can compromise the linear regression having a dramatic influence on the estimated extraterrestrial constants. The use of a relatively wide range of atmospheric airmasses is preferable.

Data from routine DS observations are used in the Brewer LP, and external calibration systems are not needed. Langley plots are performed at the different wavelengths separately, and five constants are derived.

DS measurements performed with different NDFs (see the DS procedure description in section 2) cannot be used in the same Langley plot. In [14], only measurements made with NDF 3 were used; the authors used a particular schedule to obtain a sufficient number of measurements homogenously distributed in the insolation hours. To our knowledge, few Brewer stations have used dedicated schedules, and Langley plots are generally obtained in relatively narrow intervals of airmasses.

To circumvent these difficulties, we implemented a modified LP, improving the methodology described in [14] to correct for the ozone variability and to use measurements obtained with two different NDFs.

The propagation of a UV radiation beam in the atmosphere is described by the Beer–Lambert–Bouguer law:

$$\ln I_i = \ln(T_i I_0) - m(\tau_{\text{Abs}} + \tau_{\text{Ray}} + \tau_{\text{Mie}}) \qquad (1)$$

where I_i is the measured radiance with the used NDF reduced at the mean sun–earth distance, I_0 is the radiance at the top of atmosphere at the mean sun–earth distance, or extraterrestrial constant, T_i is the transmissivity of the used NDF, m is the airmass and the terms enclosed in brackets are the optical depths due to the absorption by atmospheric gases, molecular scattering, and aerosol extinction, respectively. Equation (1) can be written as follows:

$$\ln I_i(m,\lambda) = \ln[T_i I_0(\lambda)] - m\left[2.6858\times 10^{20}\cdot D_{\text{O}_3}\cdot \sigma_{\text{O}_3}(T,\lambda) + \frac{p}{p_0}\tau_{\text{Ray}}(p_0,\lambda) + \text{AOD}(\lambda)\right] \qquad (2)$$

where λ is the wavelength, D_{O_3} is the total column ozone in Dobson units, $\sigma_{\text{O}_3}(T,\lambda)$ is the ozone absorption cross section per molecule at temperature T, $\tau_{\text{Ray}}(p_0,\lambda)$ is the Rayleigh optical depth at pressure $p_0 = 1013$ mb, and p is the atmospheric pressure; absorption by other atmospheric gases, such as SO_2 and NO_2, has been neglected.

We define the variable Y as

$$Y(m,\lambda) = \ln I_i(m,\lambda) + m\tau_{\text{O}_3} - \ln\left(\frac{T_i}{T_3}\right) \qquad (3)$$

where τ_{O_3} is the ozone optical depth and T_3 is the transmissivity of NDF 3. Combining expressions (2) and (3) we obtain:

$$Y(m,\lambda) = \ln[T_3 I_0(\lambda)] - m[\tau_{\text{Ray}}(p,\lambda) + \text{AOD}(\lambda)]. \qquad (4)$$

The ratio $\frac{T_i}{T_3}$ is determined as described by [14], i.e. from measurements performed with different filters at small time intervals; the measured total ozone (derived from the same DS measurements) is used to calculate Y. In the modified Langley plot the variable Y is plotted versus m at each wavelength. The plot intercept gives the natural logarithm of the extraterrestrial radiance, normalized at the NDF 3 values.

The value of $\ln[T_3 I_0(\lambda)]$ is calculated from all half days of the dataset. The value is considered representative of the extraterrestrial constant only if it satisfies the following selection criteria:

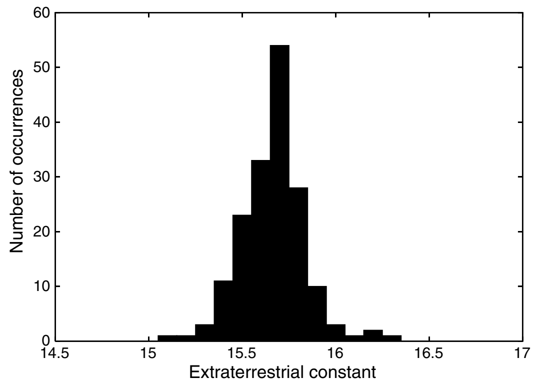

Figure 1. Distribution of the values of the natural logarithm of extraterrestrial radiances at 320.1 nm for the year 2004 in Lampedusa (Brewer 123). The median is 15.702 and the standard deviation of the mean is 0.013.

(1) the LP is obtained from at least ten direct sun measurements taken at airmass $m \leqslant 3$ (for minimizing diffuse irradiance and stray light related underestimations [23] and polarization related overestimations [28] of LP's intercept), and the standard deviation of the values of the airmass at which the radiance measurements are performed, σ_{m} must be >0.25;

(2) the standard deviation of the total ozone derived from each of the ten direct sun measurements is <2.5 DU;

(3) the root mean deviation of the measured radiances from the fitting function is <0.025.

Condition (1) is intended to eliminate all cases with few data points covering a limited range of airmasses. Condition (2) requires that the errors on total ozone are small throughout the airmass range where the LP is performed; large errors are generally associated with cloudy conditions (see i.e. [25]), which are not suitable for LP. Condition (3) sets a threshold on the goodness of the fit, and is meant to remove cases of high aerosol variability. The value of the threshold has been identified empirically, after trying different values and after visual inspection of the LPs.

The extraterrestrial constants are chosen as the median of all the determinations of $B = \ln[T_3 I_0(\lambda)]$ accepted by the selection criteria in the chosen time interval.

We analysed data from two Italian Brewer spectrophotometers: Brewer 067 operating in Rome (41.9°N, 12.5°E), and Brewer 123, operating in Lampedusa (35.5°N, 12.6°E). Brewer 067 is a mark IV type operational since 1992 [26, 27], and is located in the centre of a large urban environment. Brewer 123 is a mark III type operational since 1998 [4, 25] on the remote Mediterranean island of Lampedusa. When the median of B is computed, an appropriate time interval should be chosen, in which the extraterrestrial radiation has not changed. We studied the variability of B over the years for both stations and we found no seasonal trend. From visual inspection of the distributions of data we chose to compute the median value of B on an annual basis for the period 2002–2004 in Rome and for the whole period in Lampedusa, a three-year basis for the period 1995–1997, and a four-year basis for the period 1998–2001 in Rome. We did not analyse data for the period 1992–1994 in Rome owing to technical problems.

Figure 1 shows the distribution of the accepted values of B at 320.1 nm for year 2004 for Lampedusa. The histogram is well described by a Gaussian curve, indicating that the retrieved values have a symmetric distribution. The one-standard-deviation uncertainty on the value of the extraterrestrial constant is less than 1.3%, while the one-standard-deviation of the average is smaller than 0.1%. The uncertainty on the AOD at 320.1 nm, due to the statistical error of the extraterrestrial constant, is $\leqslant 0.015$. This value is smaller than that obtained with conventional LP of Brewer spectrophotometer data [11, 14].

Once the extraterrestrial factors are determined at each wavelength, AODs can be calculated from single DS measurements using

$$\mathrm{AOD}(m,\lambda) = \frac{1}{m}[\overline{\ln T_3 I_0(\lambda)} - Y(m,\lambda)] - \frac{p}{p_0}\tau_{\mathrm{Ray}}(p_0,\lambda) \quad (5)$$

$$\sigma_{\mathrm{AOD}}(m,\lambda) = \frac{1}{m} \times \sqrt{\sigma^2_{\ln T_3 I_0}(\lambda) + \sigma^2_Y(m,\lambda) + \left(m\frac{\tau_{\mathrm{Ray}}(p_0,\lambda)}{p_0}\right)^2 \sigma^2_p}. \quad (6)$$

Instantaneous values of the atmospheric pressure are used in expression (5). When the atmospheric pressure is not available, a fixed value for the Rayleigh optical depth is used. We used data obtained with NDF 2 and NDF 3, and the AODs are calculated only from DS measures obtained with the same two filters.

As discussed in [9, 23] and [28], various sources of systematic errors affect the AOD retrieval.

A systematic overestimate of the measured direct radiance is produced by the fraction of the diffuse irradiance entering the instrument's field of view (FOV). This effect influences the extraterrestrial constant determinations obtained with the Langley plot, as well as the AOD determination [23]. The net effect is an underestimate of the AOD. The magnitude of the effect, as pointed out by Arola and Koskela, increases with solar zenith angle and instrumental field of view, and decreases with wavelength. Moreover, the amount of diffuse light entering the instrumental field of view depends also on the aerosol properties, see e.g. [2]: in general, the larger the particles, the stronger is the forward scattering peak, and the larger the contamination of the direct from the diffuse irradiance. The intensity of the forward scattering peak depends also on the aerosol amount in the atmosphere. The Brewer field of view is 2.6°. Arola and Koskela have estimated that this effect produces a systematic underestimation of the AOD by 0.01–0.02 for an airmass limit of 4.5. Meloni *et al* [2] have estimated the effect on the AOD at 416 nm produced by the diffuse irradiance entering a field of view of 3.27° for 60° solar zenith angle, and for large particles (that produce a strong forward scattering peak, and a larger contamination of the direct radiance measurement) with an AOD of 0.4 at 416 nm. The AOD at 416 nm is underestimated by 0.02. The effect at the Brewer longer wavelengths (e.g. 320.1 nm) is expected to be similar or smaller, for relatively high numbers of large particles and the same solar zenith angle, due to the smaller field of view. In order to reduce the influence of the diffuse radiation contamination on the results we have limited our analyses to airmasses $\leqslant 3$.

A second systematic error on the AOD determination is described in [28]. This paper shows that two polarization sensitive elements in the Brewer optical system produce a different sensitivity for different polarization of the measured radiation. The effect increases with solar zenith angle. Cede *et al* found that this effect produces a 2–4% overestimation of the calibration factors at each wavelength, and a 0.01–0.04 overestimation of AOD [28]. No correction for this effect is considered in our analysis. However, the applied limit on the used airmass range helps reduce the error due to polarization. With this limit, the systematic error due to polarization effects is estimated to be <0.02.

As discussed in section 2, stray light also produces a systematic underestimate of the retrieved AOD. As discussed in [23], this effect is negligible for double monochromator Brewers, and at wavelengths $\geqslant$313.5 nm for single monochromator Brewers. As will be discussed below, we use AOD data obtained only at 320.1 nm, where the stray light effect is negligible for both Brewers mark III and mark IV.

Neglecting NO_2 and SO_2 absorption in equation (2) leads to an overestimation of the AOD. This effect is assumed to be small in Lampedusa, where both SO_2 and NO_2 column amounts are small, while it may be significant in Rome. Column amounts of both SO_2 and NO_2 (a technique to measure nitrogen dioxide total column amount by means of Brewer DS data is described in [29]) are measured by the Brewer, and a correction for this effect, although not applied in this work, is possible.

In [9], a discussion on the impact of variations of the effective ozone temperature on the AOD determinations with the Brewer is reported. This effect decreases with wavelength, due to the fast reduction of ozone absorption cross section. The effect at 320.1 nm, the wavelength used in this analysis, is estimated to be negligible.

The temporal variability in the transmissivity ratio between the used NDFs may also produce a systematic error. We have studied the variations of the relative transmissivity of NDF 2 with respect to NDF 3 and found that it has remained reasonably constant at both sites in the whole period (3.20 ± 0.60 in Rome, 4.75 ± 0.74 in Lampedusa). The quarterly means of the ratios of transmissivities always differ from the mean value by a smaller quantity than the standard deviation.

4. Results

As previously discussed, the method described in section 3 has been applied to the DS measurements of Brewer 067 (operating in Rome) and 123 (operating in the Mediterranean island of Lampedusa). We analysed data for the periods 1995–2004 in Rome, and 1998–2004 in Lampedusa.

Data from Lampedusa were analysed using three different procedures: the first one is a conventional LP, as described in [14]; the second one includes the use of NDF 2 and 3 in the conventional LP; the third one is the improved algorithm described in section 3. The same screening criteria to define acceptable LPs have been used in the three cases. Figure 2 shows the number of accepted LPs obtained during the years 1998–2004 with the three methods. In summer 2000 the D3 routine, that forces the Brewer to perform DS measurements with filter 3 [14] throughout the day, has been included in

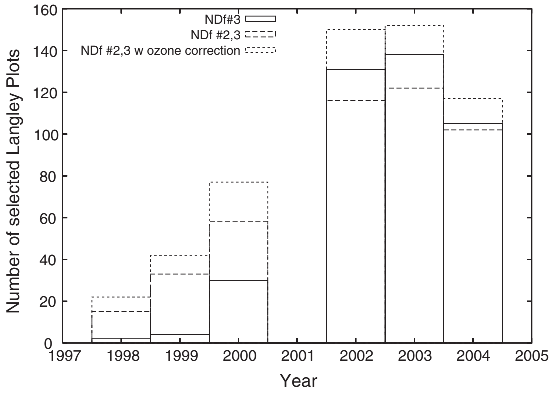

Figure 2. Statistics of the number of valid determinations of the extraterrestrial constant using standard LP and data from NDF 3 only (solid line), standard LP and data from NDF 2 and 3 (dashed line), and the improved method with modified LP and data from NDF 2 and 3 (dotted lines) at Lampedusa (Brewer 123) for the period 1998–2004.

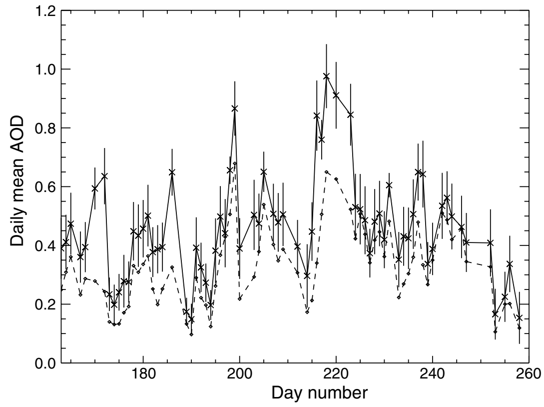

Figure 3. AOD daily means obtained from Brewer (solid line with crosses and error bars, $\lambda = 320.1$ nm) and MFRSR (dashed line with diamonds, $\lambda = 416$ nm) observations during the period 11/06–16/09 2003 in Lampedusa.

the operational schedule, and the conventional analysis uses a relatively wide range of airmasses. The use of data with two neutral density filters implies that a larger number of data points is used in the Langley plot. The reduction ($<10\%$) in the number of selected LPs is consequently due to the difficulty of obtaining a reliable LP from a larger number of observations. In years 1998 and 1999 the number of selected conventional LPs is small, due to the relatively small number of DS routines included in the operational schedule at that time, and partly to discontinuous Brewer operation. In 1998 and 1999 the use of data with two NDFs produces a significant increase in the number of selected LPs. The improved algorithm produces the largest number of selected LPs throughout the period. The improvement is critical in years 1998–2000, when dedicated routines were not implemented.

The possibility to use a larger number of LPs with the same selection criteria in principle allows a better determination of the extraterrestrial constants, and permits the investigation of possible temporal changes over relatively short time periods.

As previously discussed, the method allows the derivation of the aerosol optical depth at the five wavelengths used in the DS routine. The estimated one standard deviation error on the AOD is smallest at 320.1 nm, due to the highest signals and smallest influence of the uncertainties on ozone and Rayleigh optical depths. A 1% uncertainty on total ozone and a 5 mb uncertainty on the atmospheric pressure produce an uncertainty of 0.014 on the optical depth at 306.3 nm, 0.007 at 313.5 nm, and 0.005 at 320.1 nm. The estimated uncertainty on the AODs is larger, since measurement errors must also be considered. An aerosol layer with AOD $= 0.30$ at 320 nm and Ångström exponent $= 1.5$ has AOD at 306.3 nm equal to 0.32. The difference between AODs at 306.3 and 320.1 nm is smaller than the sum of the measurement uncertainties at the two wavelengths. Thus, the use of different wavelengths within such a small spectral range does not provide useful information on the spectral behaviour of the AOD. Consequently, we will discuss only optical depths retrieved at 320.1 nm.

The methodology has been validated by comparing the retrieved AOD values at 320.1 nm with independent measurements obtained with a CIMEL sun photometer and a MultiFilter Rotating Shadowband Radiometer (MFRSR). An MFRSR has been operational side by side with the Brewer 123 in Lampedusa since 2001. For further information about the instrument and optical depth retrieval techniques see [4, 15, 32]. A CIMEL sun photometer, operating in the frame of AERONET, is operational at Rome Tor Vergata, about 15 km south-east of Brewer 067. For further information about the instrument and AOD retrieval see [16, 31]. Owing to differences in the operative wavelengths of the three instruments (the CIMEL operates at 440 nm and longer wavelengths, while the MFRSR operates at 416 nm and longer wavelengths), and the distance between the Brewer and CIMEL in Rome, a close numerical agreement is not expected. Although the use of AODs at the same wavelengths would have been the best solution for a comparison, we used the values of AOD at the closest wavelengths among those available.

Figure 3 shows the time series of the AOD daily means derived from Brewer and MFRSR in Lampedusa during summer 2003. The daily standard deviation of the Brewer AOD is reported in the figure. The same time evolution appears in the two datasets; as expected, the AOD at 320.1 nm is larger than at 416 nm. Reference [30] discussed in detail the behaviour of the aerosol optical depth and of the aerosol properties during summer 2003. Periods characterized by a strong wavelength dependence (in most cases corresponding to forest fire aerosols during summer 2003, e.g. in particular between day number 215 and 235) and by a weak wavelength dependence (corresponding to desert dust, e.g. in the periods 180–185, 196–200, 205–211, and 235–244) of the AOD evidently appear. The alternation of these different aerosol types is consistent with the results discussed by [30, 32].

Figure 4 shows the time series of daily mean AODs derived from the Brewer and CIMEL in Rome during part of 2003. Also at this site, an agreement between the time evolutions emerges, with Brewer AODs constantly larger than CIMEL data.

Figures 5(a) and (b) show the scatter plots of daily mean AOD measurements in Lampedusa and Rome. The correlation coefficient is $R^2 = 0.93$ in Lampedusa and $R^2 = 0.94$ in Rome. The high correlation between the independent datasets

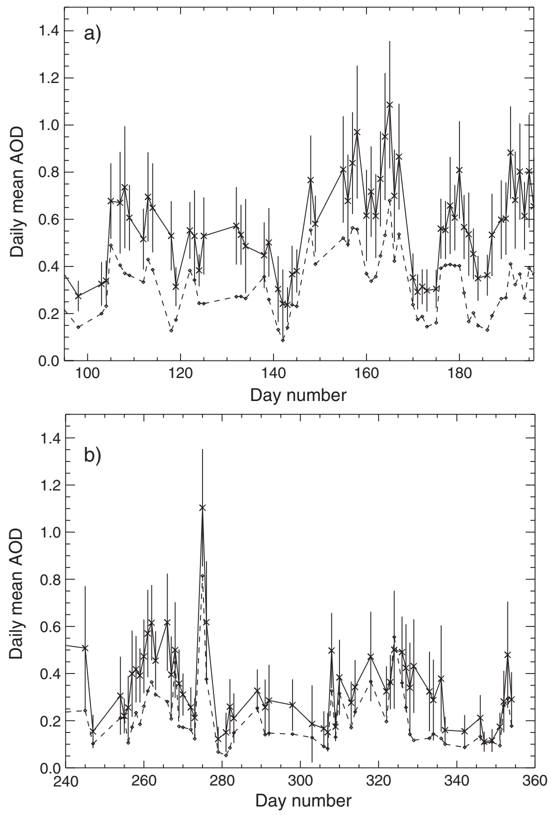

Figure 4. AOD daily means obtained from Brewer (solid line with crosses and error bars, $\lambda = 320.1$ nm) and CIMEL (dashed line with diamonds, $\lambda = 440$ nm) during the periods (a) 04/04–14/07 and (b) 28/08–26/12/2003 in Rome.

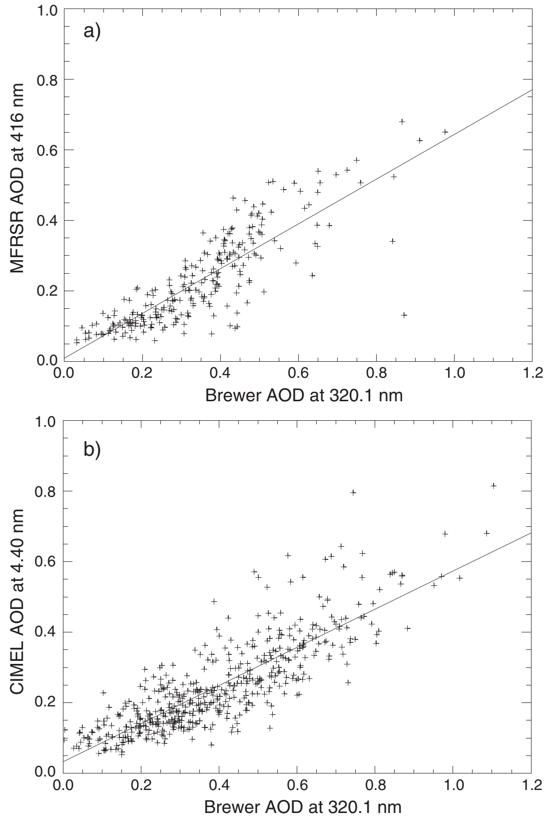

Figure 5. Scatter plots of daily average AOD values from independent datasets in (a) Lampedusa and (b) Rome.

at both stations gives an indication of the consistency of the methodology.

5. Conclusions

An improved Langley plot method has been implemented to retrieve the aerosol optical depth from Brewer direct sun measurements. The method uses measurements obtained with two different neutral density filters over a relatively wide range of atmospheric airmasses. The modified Langley plot accounts for short-term variations of total ozone. Total ozone is derived from the same direct sun measurement used in the Langley plot.

The improved algorithm has been applied to data collected with a Brewer mark IV operational in Rome, in an urban environment, and with a Brewer mark III, operational in Lampedusa, a remote island in the Mediterranean. The improved algorithm produces a larger number of reliable Langley plots than the standard Langley plot procedure. The larger number of Langley plots allows for a better statistical characterization of the extraterrestrial constant and a better study of its temporal variability.

Using the derived extraterrestrial constants, aerosol optical depths are calculated at the operational Brewer wavelengths. Daily average optical depths at 320.1 nm have been compared with independent measurements obtained with a CIMEL sun photometer in Rome at 440 nm, and with an MFRSR in Lampedusa at 416 nm. Sun photometers and Brewer aerosol optical depths show a similar evolution at both sites; the correlation between the two datasets is 0.94 in Rome, and 0.93 in Lampedusa. Brewer AODs are, as expected, consistently larger than at 416 and 440 nm.

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