Twenty years of UV aerosol optical depth measurements by a Brewer spectrophotometer over Thessaloniki, Greece

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Abstract. Direct-sun spectral UV measurements from a double monochromator Brewer spectrophotometer, operating at Thessaloniki, Greece, have been analyzed and the spectral aerosol optical depth (AOD) in the wavelength range 320 – 360 nm has been derived for the period 1997 – 2017. From the AOD spectral measurements the Angstrom exponent was calculated for the particular spectral region. These two products have been validated against the corresponding products of a collocated Cimel sun-photometer (AERONET) to ensure their good quality. Then, the short and long term variability of the AOD and the Angstrom exponent were analyzed and discussed for the entire period of study. The analysis yielded a strong negative trend in the AOD at 340 nm of the order of 0.1 per decade. This negative trend confirms previous studies in the area and over similar environments in Europe and is mainly attributed to successful implementation of air-pollution abatement measures during recent decades.

1 Introduction

Aerosols play a major role in climate formation and are one of the main uncertainty factors in global climate models. They also impact air quality, with negative effects on human health (Fuzzi et al., 2015). Estimations of the radiative forcing (Nabat et al., 2014) and the concentration of aerosols can be derived by the Aerosol Optical Depth (AOD), as measured by ground based and satellite instruments. Satellite instruments can not always provide accurate results, especially in urban environments were spacial and temporal scales are small, and thus have to be supplemented by ground measurements (Xiao et al., 2016). Understanding of AOD variability can be improved by using long-term records, of good quality data. In this paper, the variability of AOD at 340 nm and the Angstrom exponent (AE) in the range 325 – 360 nm are studied and presented for a 20-year period, using direct irradiance spectra measured by a double-monochromator Brewer spectrophotometer. A comparison between the measurements of the Brewer and a co-located Cimel sun-photometer is performed, in order to estimate the quality of the AOD derived by the Brewer.

2 Instruments, data and methods

At the Laboratory of Atmospheric Physics (LAP), Thessaloniki, Greece ($40.634°N$, $22.956°E$, $60m$ a.s.l.), a single-monochromator MKII Brewer with serial number 005 (B005) is operating since 1982, and provides automated continuous measurements of total ozone. Since 1993, a second double-monochromator MKIII Brewer with serial number 086 (B086) is also operating, next to the first one, performing direct and global spectral measurements of the solar UV irradiance in the range 290 – 363 nm with a step and a resolution of 0.5 nm. The spectral measurements of B086 are considered superior to those of B005, due to its extended spectral range (B005 operates in the range 290 – 325 nm, with step and resolution of 0.5 nm), and mainly due to its more efficient rejection of stray light (e.g. Karppinen et al., 2015). Since 1997, the direct sun spectra of B086 are calibrated using the methodology suggested by Bais (1997), and are suitable for the retrieval of the AOD in the operational spectral range of the instrument (Kazadzis et al., 2005).

At the same location, a Cimel photometer is installed as part of the Aerosol Robotic Network (AERONET). The instrument performs various measurements of aerosol optical properties, including AOD, at different wavelengths in the spectral range of 340 – 1020 nm (Dubovik and King, 2000). Here, we are using the "level 2.0" data from AERONET, as reference of comparison with the Brewer AOD.

In the context of the present study, the direct-sun spectra measured by B086 have been re-evaluated. Novel techniques have been used for the correction of the data for the effects of dead time (Fountoulakis et al., 2016b) and temperature (Fountoulakis et al., 2016a), in addition to the corrections applied in the past (Garane et al., 2006; Kazadzis et al., 2005, 2007). Then, the AOD for the period 1997 – 2017 was calculated for wavelengths 305 – 363 nm using the methodology described in Kazadzis et al. (2005), for solar zenith angles (SZAs) less than $75°$. The AE was then calculated for the range 325 – 360 nm. Shorter wavelengths were rejected because of the strong effect of ozone at these wavelengths, which increase uncertainties in the retrieval of AOD.

Kazadzis et al. (2007) estimated an uncertainty of ~0.05 in the retrieval of the AOD in UVA region (i.e. wavelengths longer than 315 nm), which is applicable in this study. The comparison of the AOD at 340 nm with the corresponding AOD from Cimel, confirms these findings. Uncertainties tend to increase at small and large SZAs (near 15° and 75° respectively). At small SZAs the effect of errors in the calibration factor become stronger, while at large SZAs uncertainties in the measurements become more important. The uncertainties in the determination of the AOD are generally larger for low values of AOD and the calculated AE cannot be considered reliable in these cases.

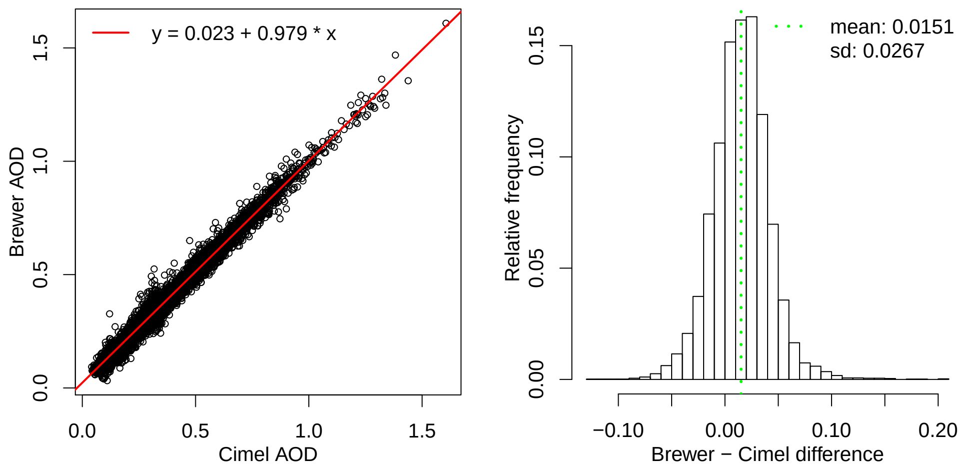
B086 performs a full spectral scan of the direct sun irradiance every ~30 minutes. The scans of the global UV irradiance (performed every ~20 minutes). are interrupted, and direct sun measurements are performed at 10 nm intervals (300 - 360 nm with a step of 10 nm). While a global irradiance scan is recorded, the direct spectral irradiance is sampled every 10 nm in the range 300-3260 nm. As a result, the temporal resolution of the direct irradiance measurements at these wavelengths is less than 15 minutes. The corresponding temporal resolution of the AOD from Cimel is ~45 minutes. A significant advantage of using the AOD data set of B086 is that it is one of the longest data sets globally, since AOD measurements in the UV from ground based or satellite instruments before 2000 are rare. In this study, only the AOD from direct-sun spectra (SS) have been analyzed and presented.

Brewer and Cimel measurements are not concurrent. In order to compare the two data sets, we applied a temporal match method, where every Brewer measurement was coupled with the closest in time value from Cimel. We restricted the maximum time difference between measurements to 7 minutes. This value was selected by inspecting the number of matched Brewer measurements, the Root Mean Square Difference and Mean Bias Error between the data sets. Our focus was to maximize the utilization of Brewer data and to be able to assume, that atmospheric conditions haven't changed significantly between measurements.

In order to remove the annual cycle of AOD and to be able to investigate the trend over longer time periods, we have applied a deseasonalization method, as follows. We use the daily AOD means to compute monthly AOD means for the whole data set (1997 - 2017). Including only months that have measurements for at least 50% of the days, we aggregated the time series by month, producing the mean AOD for each month of the year. The deseasonalized data were calculated by subtracting the corresponding AOD value from each monthly mean. We also applied the same analysis, for finer time scales and found that the variability of the data is such, that the annual cycle can not be adequately described in these scales.

3 Relation between Brewer and Cimel AOD measurements at 340 nm

The comparison of the temporally matched data of AOD at 340 nm between Brewer and Cimel (allowing time differences of less than 7 minutes) shows a good correlation. The matched data consists of 7400 coupled points, acquired over a period of 1135 days and yield a strong linear correlation factor (0.98) between the two data sets with high statistical significance ($R^2 = 0.98$), (Fig. (1a) and Table (1)). This indicates the feasibility of using Brewer measurements of AOD at 340 nm to produce a long AOD time series, that extends beyond the available Cimel measurements. Similar results have been demonstrated by Kazadzis et al. (2007) using the same data set for a shorter period. The temporally matched data, shows that Brewer tends to overestimate AOD on average by 0.015 (sd: 0.027).



(a) Linear regression AOD 340 nm. (b) Distribution of AOD difference.

Figure 1: Comparisons of Brewer (SS) and Cimel, temporal matched AOD at 340 nm. Included data have a maximum time difference of 7 minutes.

Table 1: Linear regression parameters for temporally matched AOD (430 nm) of Brewer and Cimel.

Ndata	RMSE	MAE	MAEsd	Intercept	Slope	R_squared
7400	0.0307	0.0198	0.0174	0.0231	0.979	0.983

Comparing the annual cycles of monthly means between the complete data sets, we found that Brewer measures higher AOD values on average by 0.018 (sd: 0.0076) with a maximum difference of 0.028 in March and a minimum of 0.0047 in September. The time series of the two instruments is presented in Fig. (2).

4 Comparison of Angstrom exponent from Brewer and Cimel

The two instruments perform measurements on different spectral ranges, so we can not produce an AE with the exact same spectral characteristics. We are limited to compare AE in the range 325 - 360 nm for the Brewer and 340 - 440 nm for the Cimel. We used a similar method as with AOD in order to

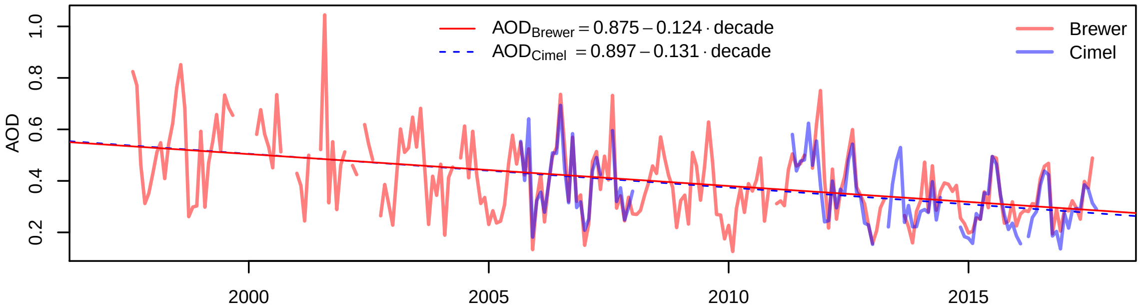


Figure 2: AOD (340 nm) monthly means time series from all data of both instruments for the period 2005 - 2017. Linear regressions are shown separately for Brewer (solid-red) and Cimel (dashed-blue) data.

temporally match the observations with maximum time difference of 7 minutes. For the calculation of the AE, the natural logarithms of AOD and the corresponding wavelengths in the region 325 – 360 nm were regressed. Then, AE is considered to be the slope of the linear fit of the two quantities. When the AOD at 340 nm is lower than 0.2, the AE is not used because of the high uncertainties in its calculation. AE values are also rejected, if the statistical significance (Man-Kendal test) of the linear fit is less than 95%.

The correlation of AE between the two instruments is not as good as for AOD (Fig. (3) and Table (2)). One reason might be the different spectral ranges of the data used for the calculation of AE. As we examined the data from Brewer, we found that AE is very sensitive to the calculation method used and the selection of spectral data. This led as to believe, that further improvement in the quality of AE is feasible.

Table 2: Linear regression parameters of the temporally matched Angstrom Exponent for Brewer (325 - 360 nm) and Cimel (340 - 440 nm).

Ndata	RMSE	MAE	MAEsd	Intercept	Slope	R_squared
9856	0.728	0.458	0.492	0.171	0.68	0.0795

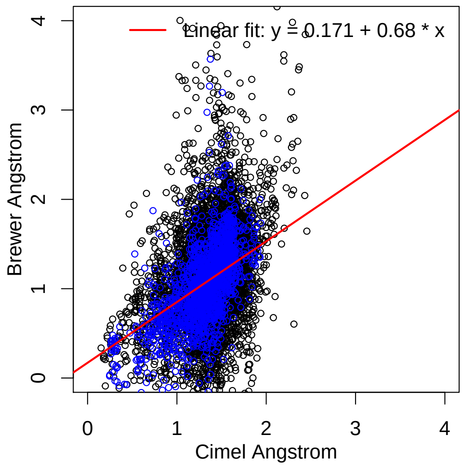


Figure 3: Linear regression of temporally matched Angstrom Exponent for Brewer (325 - 360 nm) and Cimel (340 - 440 nm). With blue are points where Brewer AOD > 0.5 . Some data point were left outside plots scope for clarity.

5 Long term AOD variability at 340 nm from Brewer

The linear regression of the monthly means of Brewer (SS) AOD at 340 nm, calculated from the daily means, shows that AOD has a rate of change of -0.13 per decade (Fig. (2) end Table (3)). The mean absolute difference between monthly fitted AOD and monthly AOD is 0.107 with a standard deviation of 0.083.

Table 3: Linear regression parameters for Brewer (SS). Slope is per decade.

RMSE	MBE	MAE	MAEsd	Slope	Slope_sde	Resid_sde	R_squared
0.136	0.115	0.107	0.083	-0.126	0.0158	0.136	0.225

6 Conclusions

The proposed methodology to derive spectral aerosol optical form Brewer direct-sun spectra, has been evaluated by comparison of AOD at 340 with data of a collocated the Cimel sun-photometer, part of AERONET. The comparison showed good agreement with an average absolute difference of 0.015±0.027. Despite the systematically higher values measured by Brewer, we were able to produce a long AOD time series, and calculate trends which are in accordance with results from other ground-based and satellite instruments (for example Floutsi et al., 2016; Provençal et al., 2017). Thus, we can propose the possibility of using Brewer, as a source of long term AOD data. For the Angstrom Exponent, we found that, despite the statistical relation of Brewer and Cimel AE, the direct comparison is hindered by the different spectral characteristics of the two instruments. Which drives us, to consider a different technique to produce AE from brewer and additional data sources for comparison in the future.

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