GEOPHYSICAL RESEARCH LETTERS, VOL. 31, L12106, doi:10.1029/2004GL019651, 2004

The fictitious diurnal cycle of aerosol optical depth: A new approach for "in situ" calibration and correction of AOD data series

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Received 5 February 2004; revised 25 April 2004; accepted 19 May 2004; published 19 June 2004.

[1] Aerosol optical depth (AOD) very often shows a distinct diurnal cycle pattern, which seems to be an artifact. This phenomenon is the result of a deficient calibration (or an equivalent effect, as filter degradation). The fictitious sinusoidal shape of the AOD diurnal cycle is a function of the cosine of the solar zenith angle (SZA) and its effect is more accentuated during mid-day. The observation of this effect is not easy at current field stations and only those stations with excellent weather conditions permit an easier detection and correction. By taking advantage of this diurnal cycle behavior because of its dependence on the cosine of the SZA, we propose an improved "in situ" calibration correction procedure. The method is named KCICLO because the determination of a constant K and the behavior of AOD as a cycle (ciclo, in Spanish). It can be seen as a modification of the classical Langley technique (CLT) with the same level of accuracy when CLT is applied at high-altitude stations, and results in an accuracy of 0.2–0.5% for the calibration ratio constant K (or 0.002–0.005 in AOD). The application of this correction method to current and old data series at sunny stations is a significant improvement over "in situ" methods, because no other information beyond the AOD data is necessary. *INDEX TERMS:* 0300 Atmospheric Composition and Structure; 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation. **Citation:** Cachorro, V. E., P. M. Romero, C. Toledano, E. Cuevas, and A. M. de Frutos (2004), The fictitious diurnal cycle of aerosol optical depth: A new approach for ''in situ'' calibration and correction of AOD data series, *Geophys. Res. Lett.*, *31*, L12106, doi:10.1029/2004GL019651.

1. Introduction

[2] The analysis of AOD diurnal variations is an important task in aerosol studies, but it is often complicated because a strong dependence exists between the measurement site and aerosol type, and variable aerosol amounts. At present, thanks to the existence of several aerosol-monitoring networks, such as AERONET (AErosol RObotic NETwork, http://aeronet.gsfc.nasa.gov) [*Holben et al.*, 1998], USDA UV-B (U.S. Department of Agriculture, Ultraviolet-B) [*Michalsky et al.*, 2001] program, WMO-GAW (World Meteorological Organization-Global Atmosphere Watch) [*Wehrli*, 2000] program, there are sufficient data series that permit adequate analysis. Few papers have been published where detailed analysis of AOD diurnal variations are shown [*Smirnov et al.*, 2002]. On the contrary, more frequently AOD variations are evaluated by means of statistical parameters (e.g., daily, monthly and yearly averages, frequency values, etc.), which lose more time-specific information. However, valuable information on AOD diurnal variation is given during intercomparison campaigns [*Schmid et al.*, 1999].

[3] While observations of a strong diurnal cycle of AOD seem to be known by researchers involved in calibration procedures, there are no relevant publications about this behaviour. On the contrary, extensive and good papers can be found about the influence of different atmospheric and instrumental parameters involved in calibration methods, where such a diurnal behaviour is recognized but not discussed in detail [*Reagan et al.*, 1986; *Korotaev et al.*, 1993; *Forgan*, 1994]. *Shaw* [1983] used a sinusoidal dependence of AOD on day time (or linear on air mass) to account for changes in atmospheric pollution or meteorological conditions. *Kremser et al.* [1984] discussed how real AOD diurnal cycles influence the Langley calibration method and hence the high errors associated with the calibration constant. Although the existence of a diurnal cycle can be due to atmospheric conditions, there is systematic evidence of calibration errors to account for this cycle. The aim of this work is to detect the existence of systematic and fictitious AOD diurnal variations and, by taking advantage of this behaviour, to implement an "in situ" correction method based only on the primitive values of the AOD.

2. Detection of the AOD Diurnal Cycle

[4] The AOD diurnal cycle behaviour has been observed at different locations and with different instruments but we restrict the discussion to our own data at "Izaña Observatory" [*Romero and Cuevas*, 2002] and "El Arenosillo" station [*Toledano*, 2003]. As a first step and as a reference, we present AOD diurnal cycle features observed at the "Observatorio Atmosférico de Izaña" (Tenerife, Canary Island, 28°N, 16°E, 2367 m.a.s.l.), because in this site this effect is clearly observed. The Observatory is located in the free troposphere and is part of the WMO-GAW network. The data were obtained with two different Sun photometers, a PFR (Precision Filter Radiometer,) and a PMOD (Physikalisch_Meteorologisches Observatorium Davos), both calibrated at the Davos Radiation Center [*Wehrli*, 2000] and the PFR belonging to the GAW network. According to *Schmid and Wehrli* [1995] and *Schmid et al.*

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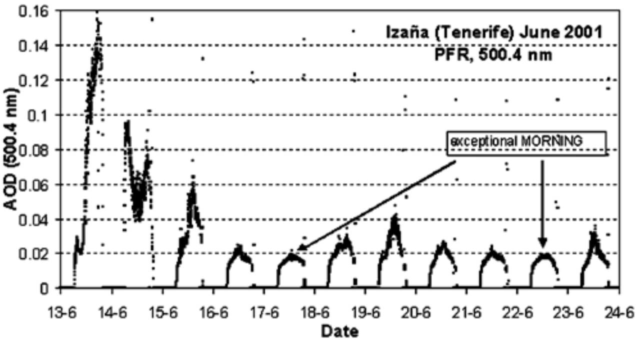

Figure 1. Diurnal evolution of PFR AOD data (for 500.4-nm) at Izaña Observatory for several days in June 2001, where the diurnal cycle is clearly observed.

[1998], the errors of calibration with laboratory lamps (about 2–4%) are larger than those obtained with the Langley procedure at high altitude stations.

[5] As the current AOD values at Izaña are very low, the observed diurnal cycle is very clear, as can be seen in Figure 1 for the 500.4-nm wavelength channel of the PFR from some days of June 2001. The AOD values approaching in this case the level of accuracy of the measurement system (1–3%). From Figure 1, the current observed AOD values using 500.4-nm filter range from 0.005 to 0.06 [*Romero and Cuevas*, 2002], when no desert dust events occur. Note how these dust events break the AOD diurnal cycle in days 13–15. Normal daily background variations are generally about 0.01–0.02, which are of the same order of magnitude as the associated absolute error for AOD [*Wehrli*, 2000; *Holben et al.*, 1998] (see also AERONET web-site). With these ranges of AOD values and taking into account these errors, the evaluation of average daily values have little or no physical or mathematical meaning.

[6] We must be sure that this effect is due to calibration and not to atmospheric or whatever effect (e.g., the effect of temperature on the detector), then atmospheric conditions and calibration procedures must be accounted for over the whole AOD data series. As an example of a detailed study over four months (June–September 2001) we present the results for a day with optimal conditions where we carry out a carefully atmospheric and calibration study, as illustrated in Figure 2. In this Figure we show the AOD evolution for the 500-nm nominal channel on 15 September 2001 for the two above mentioned Sun photometers. Here we observe a convex and fictitious shape of the diurnal cycle in the measured AOD values (squares) when the laboratory calibration constants are taken. The other curve-lines correspond to take another calibration constants as we will discuss in the results paragraph in more detail. It is obvious that the observed diurnal cycle acts to modify real daily shapes and values of the AOD and hence on data series, having important consequences on derived parameters (e.g., the Ångström α parameter).

[7] Although the usual "in situ" calibration Langley method or its various modifications [*Herman et al.*, 1981; *Forgan*, 1994, also personal communication, 2003] are a possible alternative to reprocess the AOD data, we suggest another approach in this work. This approach can be seen as a variation of the CLT, named KCICLO, because of the determination of a constant ratio K, given by the AOD cycle.

3. Methodology for Correction: "KCICLO" Method

[8] According to the Beer-Lambert-Bouguer law the AOD, τ_a, at a given wavelength λ (removed for simplicity) is given by:

$$\tau_a = \frac{\ln I_0 - \ln I}{m} - \tau_R - \tau_g \qquad (1)$$

where I is the direct solar radiation signal (irradiance, voltage, count number, etc.) and I_o is the extraterrestrial signal (the calibration constant in the case of nonabsolutely calibrated instruments); m, is the air mass and τ_R and τ_g are the Rayleigh extinction and absorption by atmospheric gases (such as ozone, water vapor, etc.), respectively. For simplicity we assume all wavelengths have no absorption. According to *Romero and Cuevas* [2002], if we use as calibration constant a value I'_o, which can be related with the true calibration constant I_o, by $I'_o = K \cdot I_o$, (where K is defined as the ratio constant), the corresponding erroneous measured AOD is τ'_a, which can be derived by:

$$\tau'_a = \frac{\ln I'_0 - \ln I}{m} - \tau_R = \frac{\ln I_0 + \ln K - \ln I}{m} - \tau_R = \tau_a + \frac{\ln K}{m} \qquad (2)$$

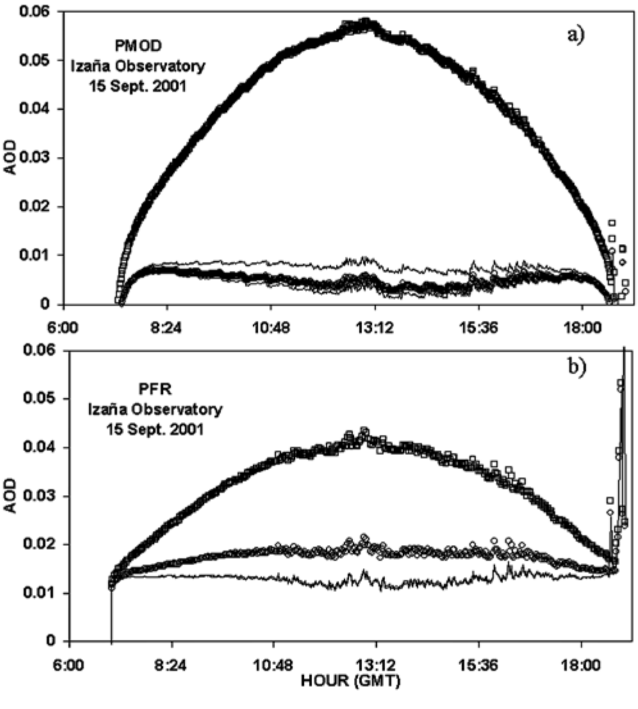

Figure 2. Diurnal AOD evolution on 15 September 2001 at Izaña Observatory (2360 m.) as measured by a) the PFR and b) PMOD Sun photometers at 500 nm (nominal) filter applying different calibration constants: the laboratory calibration (square), the Langley of the day (thin solid line), the average Langley (circles) and the KCICLO (thick solid line).

[9] Assuming as a first approximation that 1/m = cos(SZA), expression (1) gives a cosine or nearly parabolic shape of the observed false diurnal cycle of AOD values, τ'_a, if the true AOD value τ_a remains constant during the entire day (actually a half-day is sufficient for our purposes). Obviously this requirement is very hard to satisfy in the atmosphere and hence we find more or less this perfect shape. Here, we emphasize that the error of calibration is added to the real AOD values thus manifesting this behaviour. Writing expression (2) as the error of the measured AOD we have $\Delta\tau_a = \text{InK}/m$. Similar expression can be obtained by error propagation theory [*Reagan et al.*, 1986], expressed as $\Delta\tau_a = (1 - K)/m$.

[10] From this expression, we see that the error is not constant, but is modulated by 1/m, is independent of the actual magnitude of the AOD values and only depends on the ratio constant K and SZA. The error is larger at noon (when m is near 1) and lower for high SZA. An evaluation of expression (2) can be made: For instance, an error of 1% in the calibration constant, or K = 1.01, gives a maximum absolute error (m = 1) in AOD of 0.01, while 2% gives 0.02. For m = 3, this error falls to one third or 0.003 and 0.007, respectively for the 1% and 2% cases.

[11] With K defined as above, $\Delta\tau$ will be positive for $K > 1$ (I'_o greater than I_o and $\text{In}\,K > 0$). In this case, the current calibration constant is overestimated and AOD increases in the morning and decreases in the afternoon giving a convex curve shape. For $K < 1$ ($\text{In}\,K < 0$), a concave curve shape is observed for the day and the current calibration constant is underestimated. The latter case is particularly important, because negative AOD values may occur for longer wavelengths. In such cases, we are more likely to detect a calibration problem. In the former case, however, negative AOD values are not likely and we assume the overestimated data to be good data. As we have observed in our Cimel data series in "El Arenosillo", each wavelength channel has a different shape depending on its calibration and/or time degradation.

[12] This effect is quite useful not only to detect an improper calibration, but also more importantly to correct it. Traditionally, this effect has been seen as an perturbing effect for the application of Langley method due to varying atmospheric conditions [*Shaw*, 1983; *Kremser et al.*, 1984], when in many cases it was actually a calibration problem, but we can take it as a positive effect.

[13] Note that equation (2) for AOD (τ'_a) is a linear function of 1/m, with a slope of In K and the intercept being the true AOD (τ_a), which also represents the true mean daily AOD value. Therefore, we can determine the slope by a linear regression and finally a new calibration constant, if the earlier I'_o (the current precalibration constant according to AERONET protocols) was known. Then, we can evaluate the departure, reprocess the data and so on, but this is not necessary since the correction for the AOD data series is given by the same In K. Therefore, no information is necessary about a previous calibration constant and thus we can work with non-calibrated system assuming a previous-wrong calibration constant.

[14] It is easy to find the theoretical equivalence between the KCICLO method and the CLT according to equations (1) and (2). However, some theoretical differences exist, because the K parameter is obtained from the slope in the KCICLO method, while I_o is from the intercept in the CLT. In this sense our method is very similar to a modified version of the Langley method where all the points are equally weighted [*Herman et al.*, 1981; B. W. Forgan, personal communication, 2003].

4. Results and Discussion

[15] Preliminary results about the application of this method to Izaña and "El Arenosillo" stations are presented because detailed results is out of the aim of this paper (a more extended paper is in preparation). Coming back to the discussion of Figure 2 for both Sun photometer sets of AOD data, once we have observed the strong AOD diurnal cycle (squares), we can apply the calibration constants determined with the Langley plot method for this day, obtaining a flat behaviour for AOD values (thin solid line). If we now apply the mean value determined from a series of more than 80 Langley plots for fine days, we obtain a slightly convex curvature shape (circles) for PFR or concave for PMOD. Furthermore, applying the named KCICLO method we have obtained the corrected AOD values with flat behavior (thick solid line), about 0.01, showing differences between the two instruments about 0.003. These values are closed to those given using the Langley calibration constant of the day in the case of PFR but in the case of PMOD are slightly greater than the other two sets of AOD values. As can be seen the AOD values are very sensitive to changes introduced by the calibration constants. Atmospheric conditions during September 15 were very stable. Visual observations agree with balloon sounding about a strong thermal inversion layer, located at 2000 m., thus local aerosol loading can not reach Izaña altitude. This was verified by surface levels of particulate material concentration measured by a Grimm particle spectrometer located at Izaña site. The conclusion of this analysis seems to indicate that the observed systematic diurnal cycle is due to calibration.

[16] Very similar patterns as in Figure 1 were also observed in the AOD data of "El Arenosillo" station for one of the Cimel Sun photometer (number #114) deployed to this station during the period July-2000 July-2001. "El Arenosillo" is a coastal station (37.1°N, 6.7°E, 60 m.a.s.l.) of INTA (Instituto Nacional de Técnica Aeroespacial) included in AERONET that is located in the southwest of the Iberian Peninsula (Spain). Some problems were detected at the beginning of the measured period since negative values were measured at infrared filters, which seem to indicate possible calibration errors. At this type of station, the detection of this diurnal behaviour is not so easy as at high altitude stations because of the large AOD temporal variability, ranging from low-medium values, about 0.1 during normal conditions, to high values during desert dust episodes, about 0.4–0.6. However, due to very often sunny and clear days (more than 80%) the detection is not so difficult if the Cimel has an appreciable error in the calibration constant (e.g., about 2% o more). Bear in mind that the calibration uncertainty for the current field Cimel is nominally about 1–2% (minimum error), but generally may be higher depending on the delay in calibration (6 months are recommended by AERONET, but it is difficult to accomplish in practice). Due to these problems the current

data of Cimel#114 in AERONET cannot reach level 2.0, but the application of this method provides a new tool to improve the data series.

[17] In general, over a given period we must select a number of clear and stable days, given by the more perfect convex or concave behavior, to apply the method with sufficient accuracy, about 10% of days. Obviously the selected days must fulfill a set of requirements, including an air mass range, a minimum of data points for the fit, an standard deviation of the fit with a given threshold, etc., but most important is the special care about the analysis of the atmospheric conditions. After that, the determination of the individual K values (linear fits) is an easy task, and a mean $\overline{K}$ value is obtained over a given measured period, together with its associated error represented by its standard deviation, STD. An accuracy under 1% is recommended according to *Slusser et al.* [2000].

[18] This type of determination was carried out for AERONET-Cimel#114 over 23 selected days of a year period that fulfill the requirements for applying the KCICLO method (for details, see *Toledano* [2003]). The mean $\overline{K}$ value and its STD for the four aerosol filters were determined, where wavelength channels at 1020-nm and 870-nm differed from the correct calibration by 5.0% and 4.0%, respectively. The low values of STD (0.002–0.005 or 0.2–0.5%), an order of magnitude less than the correction we make for the calibration constant indicates that the method has sufficient accuracy to carry out this process of calibration correction. The errors associated with individual K values are generally smaller than the observed variability of K for different days due to the variability of the atmosphere. Also, depending on the variation over the entire period, we can choose a unique $\overline{K}$ value or various values, depending on the filter degradation, although no clear degradation trend was detected in this study case.

[19] These preliminary results seem to indicate a successful application of the KCICLO method. The KCICLO method is best suited for field stations than CLT (specially for AERONET users) because in this case we take benefit of the same effect given by 1/m, although at high altitude station both methods seem to yield very similar results.

5. Conclusions

[20] The proposed KCICLO method, based on expression (2), for correcting AOD data is a significant contribution for "in situ" calibration methods. The resulting corrections yield a level of precision for the correction of 0.2–0.5%, a level that is difficult to achieve by other methods at most current field stations. The method is based on the detection of a diurnal cycle in AOD and takes advantage of this effect given by the linear dependence of the AOD on the inverse of the air mass. This method does not replace the Langley or other methods but its application seems to be more operational at field stations than other procedures because only AOD data are necessary (it can also improve current Langley procedure). The requirement of a sufficient number of clear days, as the KCICLO method needs, is a drawback for many stations and needs to be augmented by other methods. However the detection of this systematic diurnal cycle (being sure that it is not due to atmospheric conditions) is a clear indication of the possibility of application of the method at this site. We emphasize that the error due to intrinsic calibration procedures for the current field instrumentation for AOD studies is generally greater than 1%. This error produces a false diurnal AOD cycle, which makes the detection of trends difficult at background remote sites (e.g., Izaña), where the AOD values are of the same magnitude of these errors.

[21] **Acknowledgments.** We thank the Esat-INTA, AERONET and PHOTONS (PHOtométrie pour le Traitement Opérationnel des Normalisation Satellitaire) teams for their technical support and advice. Specially we thank Dr. Nels Laulainen from PNNL (Pacific Northwest National Laboratory), Richland WA, for his suggestions and improved text. We also thank Spanish Commission for Research and Technology (CICYT) for the financial support.

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