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On Langley plots in the presence of a systematic diurnal aerosol cycle centered at noon: A comment on recently proposed methodologies

Franco Marenco[1,2]

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[1] This short paper contains a discussion on the use of Langley plots for Sun photometer and Brewer spectroradiometer calibration in the presence of a diurnal aerosol cycle. It is shown that with Langley plots alone, it is impossible to correctly identify or remove atmospheric variations having a 24-hour periodicity with an extreme at local noon. Therefore experimentalists must either be able to exclude such a periodicity using additional measurements, or approach different calibration techniques such as the approximation by linear sections.

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1. Introduction

[2] Langley extrapolation can be used to determine the extraterrestrial constant of Sun photometers, or in other words measure the solar irradiance I_0 at the top of the atmosphere with ground-based instrumentation [*Ångström*, 1970; *Shaw et al.*, 1973]. With this method, the effect of atmospheric attenuation is accounted for by observing the variation of the ground-level direct-Sun radiance I when the air mass factor $m \simeq 1/\cos\theta$ varies during a morning or afternoon (θ is the solar zenith angle). On the assumption of a constant atmospheric optical depth τ, the plot of (ln I) versus m is a straight line, and its y-intercept represents (ln I_0):

$$\ln I = \ln I_0 - m\tau. \qquad (1)$$

However, the contrary is not necessarily true: obtaining a straight line in the plot of (ln I) versus m does not allow one to conclude that τ is constant, nor that the y-intercept is (ln I_0).

[3] Recently, the Brewer community renewed its interest in the Langley method for calibration of ultraviolet direct-Sun measurements [*Bais*, 1997; *Kazadzis et al.*, 2005]. Langley extrapolation plots have been traditionally performed at high mountain sites, to minimize the influence of atmospheric variabilities typical of the Planetary Boundary Layer [*Shaw*, 1976, 1983]. Of course atmospheric variability occurs at high altitude as well, but placing the observation station above the aerosol layer near the surface greatly reduces the magnitude of interferences. Some authors have proposed that atmospheric disturbances may be averaged out if a large number of Langley plots is available [*Harrison and Michalsky*, 1994] and that in this way extrapolation may be performed at low-altitude sites [*Marenco et al.*, 2002; *Cheymol and De Backer*, 2003]. Moreover, *Silva* [2006] has proposed some simple criteria in order to reject single Langley plots affected by atmospheric disturbance:

[4] 1. Linear regression parameters of the (ln I) versus m relationship must be "good"; thresholds on the correlation coefficient, the root-mean-square and the null hypothesis probability must be respected. These criteria are called in that reference "initial statistical criteria."

[5] 2. The optical depth obtained by linear regression, τ', must match the optical depth determined by directly applying Beer's law, τ'' (in the mentioned paper a tolerance is set to twice the standard error).

[6] Performing Langley plots in the Planetary Boundary Layer exposes one's measurements to diurnal cycles, specially in noncloudy scenes where the thermal excursion at the ground can be large. Of course, the assumption of a constant atmospheric optical depth τ can be reduced to a constant aerosol optical depth if other contributions (due to Rayleigh scattering and gaseous absorption) can be determined and corrected for. *Arola and Koskela* [2004] briefly discuss the effect of a diurnal ozone cycle on the Brewer spectroradiometer calibration and suggest that, if the ozone column concentration is known, its variation be accounted for as by *Cheymol and De Backer* [2003]. In the present paper, I shall discuss the case of a diurnal cycle centered at noon for an optical depth component, such as aerosol particles, which has an unknown concentration: therefore its variation cannot be corrected for. It is necessary to remind that the possible presence of such a cycle poses a serious limit to the proposed methodologies, as was already pointed out by *Shaw* [1976]. In fact, in the presence of optical depth variations with a 24-hour periodicity and a maximum or minimum at local noon, the methods based on Langley extrapolation alone do not yield satisfactory results

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[1] Physics Department, University of Genova, Genova, Italy.

[2] Also at Istituto Nazionale di Fisica Nucleare, Genova, Italy.

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capable of correctly determine I_0 or at least detect the atmospheric disturbance.

2. Mathematical Representation of the Problem

[7] The solar zenith angle θ and the air mass factor m depend upon the observer's latitude ϕ, the Sun's declination δ and the Sun's local hour angle ω, as given by *Iqbal* [1983]:

$$\frac{1}{m} \simeq \cos\theta = \sin\delta\sin\phi + \cos\delta\cos\phi\cos\omega = S + C\cos\omega, \quad (2)$$

where $S = \sin\delta\sin\phi$ and $C = \cos\delta\cos\phi$ will here be assumed constant during one measurement day (on Earth: $-0.4 \leq S \leq 0.4$; $0 \leq C \leq 1$).

[8] We now consider optical depth variations which present a 24-hour periodicity, with a maximum or minimum for $\omega = 0$ (local noon):

$$\tau = \tau_0 + \tau_1\cos\omega, \quad (3)$$

where τ_1 represents the amplitude of the diurnal variation. Substituting into equation (1), and using simple algebra:

$$\begin{aligned}\ln I &= \ln I_0 - m\tau = \ln I_0 - \frac{\tau_0 + \tau_1\cos\omega}{S + C\cos\omega} \\ &= \left(\ln I_0 - \frac{\tau_1}{C}\right) - m\left(\tau_0 - \tau_1\frac{S}{C}\right) = \ln I'_0 - m\tau', \end{aligned} \quad (4)$$

where both $(\ln I'_0) = (\ln I_0) - \tau_1/C$ and $\tau' = \tau_0 - \tau_1 S/C$ are constants. It follows that the resulting Langley plot is a straight line, indistinguishable from the constant optical depth case; however both I'_0 and τ' differ from the actual values I_0 and τ. Therefore verifying linear regression parameters, such as for instance the initial statistical criteria of *Silva* [2006], does not help excluding such a case. Nor does it help splitting the Langley plot into smaller air mass factor intervals and verifying that they all yield the same I_0, as suggested by *Kuester et al.* [2003].

[9] Let us now examine the feasibility of an additional verification: as in the work by *Marenco et al.* [2002], let us perform a postcalibration check in order to see whether τ has really been constant during the Langley plot. We therefore verify whether the optical depth derived from Beer's law, τ'', determined with the erroneous extraterrestrial constant I'_0, has varied during the measurements:

$$\tau'' = \frac{1}{m}\left[\ln I'_0 - \ln I\right] = \tau - \frac{\tau_1}{mC} = \tau_0 - \tau_1\frac{S}{C} = \tau'. \quad (5)$$

It results that not only the optical depth τ'' determined with the erroneous extraterrestrial constant I'_0 does not vary with time (while the true optical depth τ varies according to equation (3)), but that it is exactly equal to the optical depth derived from the Langley plot itself, τ'. This means that such an anomaly would not even be detected with the additional condition introduced by *Silva* [2006], which sets a limit on $\tau'' - \tau'$.

3. Random and Systematic Errors

[10] As shown in equation (4), the error on $(\ln I_0)$ is equal to $-\tau_1/C$. As during the year C only varies by ±4.3% around its average (at a given latitude), we will assume for simplicity that it is constant. In the case of variations of τ_1 from one day to the next, the diurnal cycle condition manifests itself by a spread on $(\ln I_0)$ determinations performed on different days. If some days exhibit a diurnal cycle with a maximum at noon (positive τ_1) and others with a minimum (negative τ_1), the error on $(\ln I_0)$ can in part be compensated by averaging over a large number of Langley plots. We would therefore be in the presence of a random error, and the trace left in the data by the presence of diurnal cycles would be a large standard deviation on $(\ln I_0)$.

[11] However, there is no reason to assume that τ_1 averages to zero, and even for a large set of Langley plots its average may well be larger than its standard deviation: it actually depends, for each measurement site, on the cycles for aerosol sources and local meteorology. A diurnal cycle can be, for instance, expected in an urban scenario, due to trapping and buildup of aerosols in the morning under a capping temperature inversion, with breakup and dispersal in the afternoon, due to heating and convection [*Shaw*, 1976, 1983]. If this is the case, we have a systematic error which cannot be deduced or quantified from the collected data set alone. *Shaw* [1983] reports that for this reason calibrations at Tucson, Arizona, showed extraterrestrial constants systematically lower by $\simeq 1-3\%$ with respect to calibrations at Mauna Loa Observatory (3397 m amsl). Even at the latter high-altitude site, upflowing air could contaminate results in the late morning and in the afternoon: the continuous monitoring of the particle number concentration was thus reported necessary during Langley extrapolation.

[12] Suppose that the average τ_1 (which is unknown to the experimentalist using the Langley method alone) is different than zero; in that case we end up with the following relative error on the extraterrestrial constant:

$$\frac{\delta I_0}{I_0} = \frac{I'_0 - I_0}{I_0} = e^{-\tau_1/C} - 1 \simeq -\frac{\tau_1}{C}, \quad (6)$$

and the following absolute error on subsequent aerosol optical depth determinations using Beer's law:

$$\begin{aligned}\delta\tau &= \tau'' - \tau = \frac{1}{m}\left[\ln I'_0 - \ln I_0\right] = \frac{1}{m}\ln\left(1 + \frac{\delta I_0}{I_0}\right) \\ &\simeq \frac{1}{m}\cdot\frac{\delta I_0}{I_0} \simeq \frac{\delta I_0}{I_0}(S + C\cos\omega), \end{aligned} \quad (7)$$

and therefore

$$\delta\tau = -\frac{\tau_1}{mC} \simeq -\tau_1\left(\frac{S}{C} + \cos\omega\right). \quad (8)$$

Note that this error varies with time, exhibiting a constant part plus a diurnal cycle.

[13] This far, the case of a diurnal cycle expressed by equation (3) has been discussed, i.e., a smooth function having an extreme at noon, which has been represented here with a cosine law, and *Shaw* [1976] represented with a parabola about the noon point (following Taylor series expansion). As a matter of fact, it is credible that many types of smooth monotonic optical depth variation during a

half-day can result in linear, and thus "good" Langley plots, with the result of overestimation or underestimation of the extraterrestrial constant even if the cosine law is not strictly respected. This is specially true if Langley plots cover a restricted air mass factor interval after application of data exclusion criteria (which are necessary). In this respect, the stringent requirement introduced by *Silva* [2006] that $\tau'' - \tau'$ must be smaller than a threshold may help exclude part of such cases. This is exemplified in Figure 1 and 2 of *Arola and Koskela* [2004], where an apparently linear Langley plot leads to a nonconstant optical depth when the deduced calibration constant is applied to the same data for determining τ'' using Beer's law.

[14] If the systematic diurnal variability is asymmetrical around local noon, the morning and afternoon Langley plots may result in errors on I_0 which are opposite in sign and can in part be compensated. A purely asymmetrical pattern, for instance, is the case of a steady increase or decrease from the morning to the evening: a systematic difference between morning and afternoon Langley determinations of I_0 would reveal such an asymmetrical cycle. It would therefore be suitable, if made possible by local meteorological patterns, to include both morning and afternoon plots in the data set of Langley determinations of the extraterrestrial irradiance, and critically revise the results; but this would only reveal disturbances which are asymmetrical around noon, whereas symmetrical components would not be revealed.

[15] In general the diurnal cycle can be intermediate between being purely symmetrical and purely asymmetrical about noon: the maximum or minimum may occur during the morning (or the afternoon). In the latter case, nonlinear Langley plots would be found for the morning (afternoon), while only afternoons (mornings), which would present a monotonic optical depth behavior, can have a chance to pass linearity criteria and be considered for Langley extrapolation; they would however yield an altered extraterrestrial constant I_0' as their result. Other intermediate cases can occur with an optical depth extreme at local noon but different slopes for the morning and afternoon, as shown by *Terez and Terez* [2003, Figure 1]; in that case, again, averaging the morning and afternoon results does not solve the problem.

4. Discussion and Conclusions

[16] An intrinsic limit of the Langley method, already identified by *Shaw* [1976], has been recalled and reformulated using a simple formalism: in the presence of a diurnal cycle centered at noon, an erroneous extraterrestrial constant I_0' is obtained, and this case is indistinguishable from a constant optical depth condition. The problem arises with low-frequency optical depth variations (24-hour period), whereas higher-frequency components would easily be detected because they would alter the linearity of Langley plots. This limitation has been underestimated in the description of several methodologies proposed in the recent literature: if I_0' is taken as the extraterrestrial constant of the instrument, this affects all subsequent optical depth determinations obtained using Beer's law by a systematic error equal to $-\tau_1/mC$.

[17] *Smirnov et al.* [2002] report the diurnal aerosol variability for a number of measurement stations, and indeed for a few of them an aerosol variation centered at noon is systematically found: this is the case for instance of the town Ilorin, in Nigeria (minimum at noon) and for two maritime stations, Rottnest Island and Tahiti (maximum at noon). However, major urban sites present asymmetrical daily cycles: for instance in suburban Washington, DC, and Mexico City aerosol optical depth steadily increases from the morning to the evening; in Santiago de Chile the opposite is observed. A number of sites under urban or industrial influence show a pattern similar to Mexico City, though smaller in amplitude. For a number of biomass burning sites in Zambia, the aerosol optical depth has a minimum during the morning. Finally, no significant cycle is reported for a large number of stations, mainly for those under the influence of desert dust, but also for some maritime sites, and some stations placed in biomass burning scenarios that are distant from sources.

[18] To quantify the error committed by ignoring the possible influence due to a diurnal cycle centered at noon, let us assume $C \simeq 0.7$ (corresponding approximately to a latitude $\phi = \pm 45°$). Concerning cycle amplitude, *Smirnov et al.* [2002] report values ranging 0.01–0.2 approximately, depending upon site. Thus we may assume an intermediate optical depth amplitude $\tau_1 \simeq 0.05$, with a resulting systematic error on I_0 of -7%. The systematic error on subsequent aerosol optical depth measurements made using Beer's law would therefore be $-0.07/m$.

[19] In conclusion, the methods proposed in the literature to either average out [*Marenco et al.*, 2002; *Cheymol and De Backer*, 2003] or eliminate [*Silva*, 2006] atmospheric effects are effective only with random aerosol variations, but may fail when a component of these is repeated day after day. This means that if an experimentalist wishes to calibrate a Sun photometer or a Brewer spectroradiometer at sea level and does not have an independent knowledge of the optical depth cycle for his observational site, he must use more elaborate techniques than Langley extrapolation to deal with atmospheric variations. These techniques can be based, for instance, on circumsolar radiance measurements, such as proposed by *Tanaka et al.* [1986] and *Nieke et al.* [1999], or on the approximation by linear sections introduced by *Terez and Terez* [2003]. The methodology introduced with the latter paper is an algorithm based on the hypothesis that optical depth is a smooth function of time, releasing the constant atmosphere assumption of the Langley method; and it yields a solution for the extraterrestrial constant I_0 which is free of the type of systematic errors that has been discussed in this paper. The approximation by linear sections seems robust and may become an interesting complement to Langley extrapolation; hopefully its practical use by a number of experimentalists will reveal how well it works under different conditions.

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F. Marenco, Physics Department, University of Genova, I-16146 Genova, Italy. (tel.: +39-091-7824133)