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Atmospheric Measurement Techniques
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Supplement of

Recovering long-term aerosol optical depth series (1976–2012) from an astronomical potassium-based resonance scattering spectrometer

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S. Supplement overview

The Supplementary Information provides: (i) example plots of photometric signal (R+L) and the solar radial velocity in order to detect outliers in these two magnitudes, which are used to perform cloud-screening (S1); (ii) a detailed analysis of the dependence of aerosols content on Langley calibration procedure in order to justify the approach used to obtain extraterrestrial constants with the Mark-I during periods with relatively high aerosol optical depth (0.1<AOD<0.3), such as the post-Pinatubo eruption period and Saharan dust events (S2).

S1. Cloud screening

In the present paper we have used only Mark-I cloud-screened data. Meanwhile raw data consist of a one-second measurement on the lefthand side of the line (L) followed by a second on the right hand (R), the processed information (Mark-I Level 4 data) used in this study is calculated by finding its average and standard deviation on the basis of blocks of 42 s (40 s from 1984 on), which, when calibrated to velocity, results in an error of ≈1 m s^{-1}. This data set also includes the ratio (r). It is a quite robust quality assurance process in which each erroneous data is removed, either by cloud contamination or by instrumental errors. The counts measured by this instrument usually spans from 0.1 to 1 × 10^6 counts per second depending on sky transparency, presence of thin cirrus and mirror cleanliness. Cloud screening, as a part of the instrument's quality assurance, is a careful process of cloud detection and data removal performed day by day by the astronomer in charge of the instrument. It consists in the analysis of the photometric signal (R+L) and the solar radial velocity in order to detect outliers in these two magnitudes (see Fig. S1). In particular, solar radial velocity is a magnitude highly sensible to the presence of clouds because we know perfectly that the rotation of the Sun (viewed as a star) is averaged to zero and therefore if any cloud (low, medium or even small cirrus) are present in the sky in the path between the Mark-I and the sun, this velocity will be far from zero. The result is an effective technique to detect clouds, as it can be seen from Fig. S2.

It is also worth mentioning the scientific relevance of the Mark-I database in astrophysics, being the results extracted by this instrument using this methodology to screen clouds published in prestigious journals like García et al., 2007 or Roca-Cortés and Pallé, 2014.

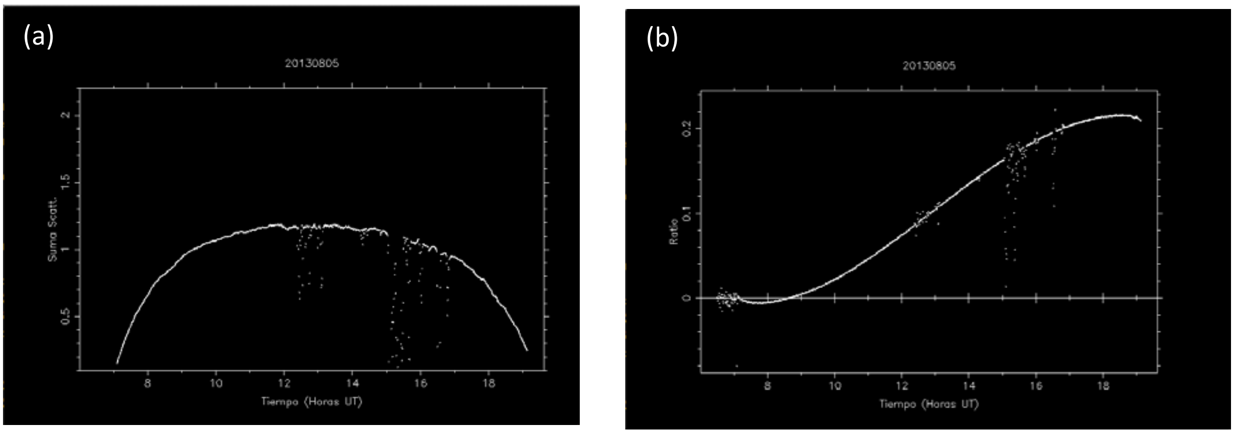

Fig. S1: (a) Photometric signal and (b) ratio extracted from Mark-I for 5 May, 2013 as case example. Outliers caused by the presence of clouds are easily identified.

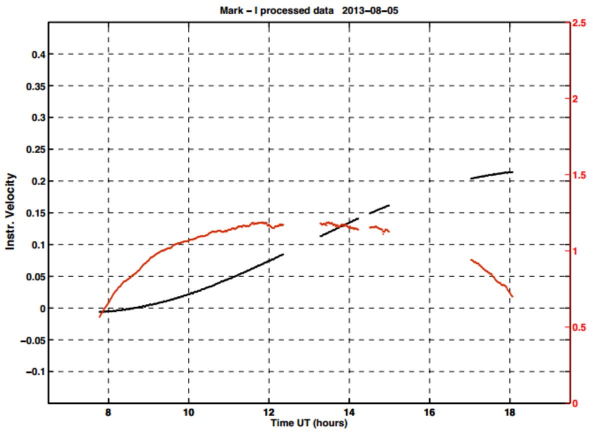

Fig. S2: Mark-I processed level 4 (cloud-screened) data for the same case example as in Fig. 1.

S2. Dependency of aerosol loads on Langley calibration procedure

The Izaña station, with an average altitude of 2370 m a.s.l., is normally located above the persistent and strong temperature inversion layer associated with the top of the marine boundary layer and with the trade wind layer, typical of subtropical regions, preventing the upcoming pollution from the urban settlements located in lower parts of the island, as well as the presence of AOD diurnal cycle at the station driven by valley-to-mountain breezes. Furthermore, the location of the Izaña in the vicinity of the most important world dust source (the Sahara) makes it to be a suitable site for aerosol, and specifically, mineral dust monitoring. In this sense, a frequent long-range dust transport above trade wind inversion layer is observed from early summer to early autumn, when the station is located within the Saharan Air Layer (SAL). This is a relatively dry and warm well-mixed layer characterized by a relative diurnal stability of aerosol optical properties. Smirnov et al. (1998) reported that a relative diurnal stability of Saharan dust optical properties has been observed during dust outbreaks at Izaña Observatory. As a result, two opposite atmospheric regimes may occur at the station:

clean background conditions, with very low AOD and relatively high Angstrom Exponent (AE) and dusty events, with relatively high AOD and very low AE since practically the entire aerosol content consists of very coarse mineral dust particles. The first one implies quite stable and pristine skies suitable to perform accurate Langley calibrations following the standard Langley plot procedure (i.e. Forgan , 1994). Some authors have developed new calibration methods to derive aerosol AOD under extremely hazy atmospheric conditions (i.e., Lee et al., 2010) and even at near-sea-level sites (Chang et al., 2014). Furthermore, other authors (Russell et al., 1993) has considered suitable to perform a Langley analysis those conditions existing after the Mt. Pinatubo eruption, with high and daily stable AODs.

Marenco (2007) demonstrated that even "good" Langleys, which can appear quite linear, might still be subject to random noise, likely caused by the variation of the AOD during the time that the Langley regression takes place. So, stability in AOD is a critical condition to obtain a good V_0. On the other hand, Kreuter et al. (2013) showed that Langley calibrations can be significantly improved by reducing diurnal variations of aerosol Angstrom parameters.

All these circumstances are present simultaneously at Izaña during Saharan mineral dust intrusions: very stable values of AOD and AE, with a nearly flat spectral response of AOD (AE values <0.3). We claim that the instrument calibration can be performed at Izaña under a priori non-ideal conditions (with relatively high AOD) following the classical Langley technique approach, in which the V_0 is inferred by extrapolating to zero air mass. Of course, this is a consequence of the special and specific characteristics of the SAL (vertical mixing and stability) and the position of the Izaña station within the SAL, and this assertion cannot be extrapolated to other sites in the world. As we have mentioned before, similar conditions (high stability in AOD and AE) may be applicable to the period of relatively high AOD values registered globally after the eruption of Pinatubo.

We present in Fig. S3 the Langley plots performed at Izaña using AERONET raw data for 15 days in 2012, differently affected by atmospheric aerosols. They correspond to data measured with air masses between 2 and 5 within the period from March to October, 2012. We have differentiated four AOD intervals for Langley analysis, ranging from AOD > 0.3 (high turbidity, in orange) to AOD < 0.1 (low turbidity, in light blue). Fig. S3 clearly shows that extrapolation to zero air mass by Langley plots for the different intervals of AOD give a nearly consistent extraterrestrial constant throughout the study period, though with some small errors. Regression analysis showed high stability in the V_0 values obtained ($V_0^{Langley}$). We can see in Table S1 how relative differences with the V_0 value used in AERONET for level 2.0 data (V_0^{AE}) is up to 0.31% for dusty events (AOD > 0.3) meanwhile values lower than 0.03% were found in case of clean events (AOD < 0.1). According to Eq. S1, these small errors yield to an absolute error on AOD from 0.003 to 0.001, lower than the precision expected for Cimel AERONET instrument, even in the case of Cimel masters. The authors want to highlight the small values of the coefficient of variation (CV) retrieved, below 0.2% in all cases. These values are similar to those obtained by Holben et al. (1998) for reference Cimel instruments at Mauna Loa (between 0.14% and 0.40%).

$$\Delta(\tau) \sim \frac{1}{m} \cdot \frac{\Delta V_0}{V_0} \quad \text{(S1)}$$

A subsequent analysis focused on AOD discrepancies between the aerosol loads obtained using both calibration approaches is shown in Table S2, where a good agreement was obtained between AOD values computed using our daily Langley calibration procedure and those AOD values extracted from AERONET. Taking into account the expected precision of the Cimel's masters (0.005-0.009, Eck et al., 1999), both Mean Bias (MB) and Mean Absolute Error (MAE) are within this precision range. Only a Root-Mean-Square-Error (RMSE) value > 0.11 was found for Langleys performed under AOD > 0.3 conditions. Considering this analysis, we can assure the suitability of those Langleys calculated at Izaña for 0.1<AOD ≤ 0.3 provided that AOD was quite stable during the Langley regression.

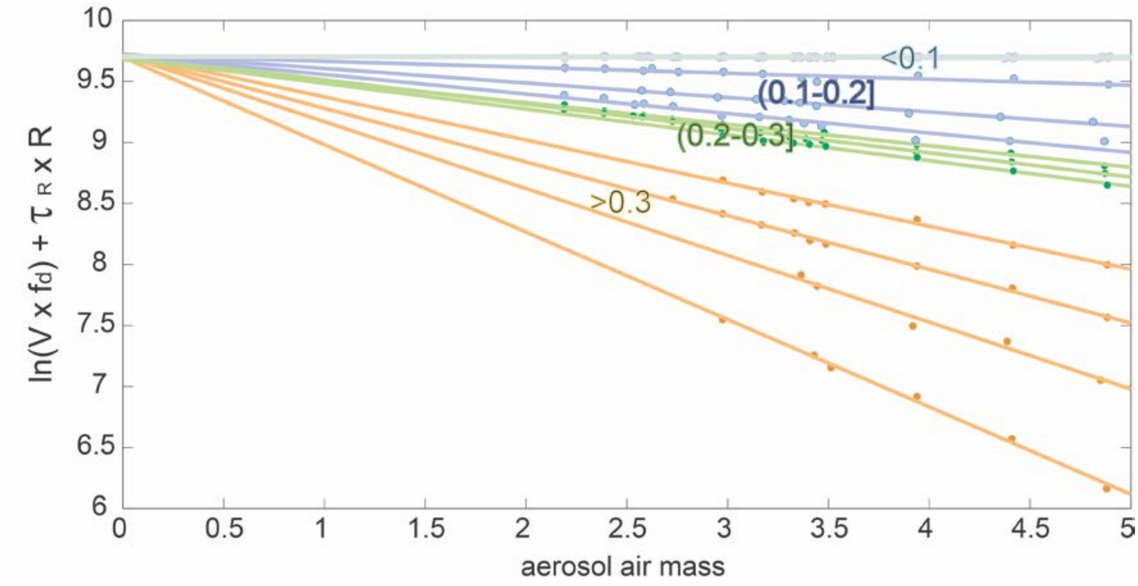

Fig. S3: Langley-plot performed at Izaña for different aerosol content intervals, from AOD>0.3 in orange to AOD<0.1 in light blue.

	DAY	$V_0^{Langley}$	V_0^{AE}	ϵ_{rel} (%)	r
AOD<0.1 **(CV=0.01%)**	May 23	9.709	9.710	0.008	0.997
	June 10	9.708	9.710	0.022	0.999
	July 27	9.709	9.709	0.003	0.999
	Sept. 3	9.711	9.708	-0.026	0.996
	Oct. 8	9.707	9.707	0.004	0.991
AOD ϵ (0.1-0.2] **(CV=0.10%)**	March 19	9.721	9.708	-0.138	0.999
	July 10	9.695	9.709	0.148	0.998
	Oct 10	9.710	9.707	-0.031	0.992
AOD ϵ (0.2-0.3] **(CV=0.09%)**	July 10	9.695	9.709	0.148	0.999
	Aug. 13	9.712	9.709	-0.031	0.999
	Aug. 15	9.716	9.709	-0.075	0.988
AOD > 0.3 **(CV=0.18%)**	April 18	9.717	9.709	-0.084	0.994
	June 27	9.680	9.710	0.306	0.993
	June 28	9.718	9.710	-0.090	0.999
	June 29	9.725	9.710	-0.163	0.999

Table S1: V_0 and main statistics of the Langley method performed at Izaña station ($V_0^{Langley}$) in comparison to AERONET (V_0^{AE}).The relative error in % (ϵ_{rel}), the coefficient of variation (CV, standard deviation divided by the mean), as well as the regression coefficient (r) have been used in this analysis.

AOD range	MB	MAE	RMSE
AOD<0.1	0.001	0.001	0.001
AOD ϵ (0.1-0.2]	0.001	0.005	0.007
AOD ϵ (0.2-0.3]	0.001	0.005	0.006
AOD > 0.3	0.001	0.008	0.011

Table S2: Skill scores for AOD comparison between computed values by using daily Langleys ($V_0^{Langley}$) and AOD values extracted from AERONET (V_0^{AE}). MB is the Mean Bias, MAE is the Mean Absolute Error and RMSE is the Root-Mean-Square-Error.

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