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Recovering Long-term Aerosol Optical Depth Series (1976–2012) from an Astronomical Potassium-based Resonance Scattering Spectrometer

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Abstract

A 37 year long-term series of monochromatic Aerosol Optical Depth (AOD) has been recovered from solar irradiance measurements performed with the solar spectrometer Mark-I, deployed at Izaña mountain since 1976. The instrument operation is based on the method of resonant scattering, which presents a long-term stability and high precision in comparison to other instruments based on interference filters. However, it has been specifically designed as a reference instrument for helioseismology, and its ability to determine AOD from transmitted and scattered monochromatic radiation at 769.9 nm inside a potassium vapor cell in the presence of a permanent magnetic field is evaluated in this paper. Particularly, the use of an exposed mirror arrangement to collect sunlight as well as the Sun-laboratory velocity dependence of the scattered component introduces some inconveniences when we perform the instrument's calibration. We have solved this problem using a quasi-continuous Langley calibration technique and a refinement procedure to correct for calibration errors as well as for the fictitious diurnal cycle on AOD data. Our results showed that calibration errors associated to the quasi-continuous Langley technique are not dependent on aerosol load, provided aerosol concentration remains constant throughout the day. It assures the validity of this technique for those periods with relatively high aerosol content required to calibrate the scattered component. The comparative analysis between the recovered AOD dataset from Mark-I and collocated quasi-simultaneous data from Cimel AErosol RObotic NETwork (AERONET) and Precision Filter Radiometer (PFR) instruments showed an absolute mean bias ≤ 0.01 in the 11 year and 12 year comparison, respectively. High correlation coefficients between AERONET/Mark-I and PFR/Mark-I pairs confirmed a very good linear relationship between instruments, proving that recovered AOD data series from Mark-I can be used together PFR and AERONET AOD data to build a long-term AOD data series at Izaña site (1976-now), suitable for future analysis of aerosols trends and inter-annual variability. Finally, the AOD preliminary trend analysis in the 29 year period from 1984 to 2012 with Mark-I AOD revealed no significant trends.

1 Introduction

Long-term measurements are the most important approach for detection of changes in atmospheric composition caused by either variation in natural or anthropogenic emissions, as well as in atmospheric processes and sinks (Collaud Coen et al., 2013).

There is a large number of publications in the literature aimed at analyzing long-term trends in variables such as insolation (Wild, 2009), solar irradiance (Dutton and Bodhaine, 2001), temperature (Jones and Moberg, 2003; Wild, 2009) or aerosol burden (Ruckstuhl et al., 2008; Nyeki et al., 2012; Collaud Coen et al., 2013; Nabat et al., 2013) in order to reveal the spatial and temporal variability of Earth's climate and its atmospheric composition. This information is crucial in the diagnosis of current and past climate as well as in the projections of future climate change since long-term records enable the development and enhance the skills of atmospheric numerical models through our theoretical understanding of radiative processes.

Such studies have shown the significant radiative impact of volcanic eruptions on at-ground solar irradiance (Dutton and Bodhaine, 2001). The eruption of Mt. Pinatubo in June 1991 is an example of a drastic reduction in direct normal irradiance (from 25–30 %), greater than that observed after El Chichón eruption in 1982, leading to a nearly global but non-uniform tropospheric cooling $\approx 0.5\,°C$ (Dutton et al., 1992). Anomalies caused by volcanic eruptions used to be the most visible pattern to identify in a long-term record, introducing an important disturbance in trends analysis that must be taken into account. Other studies focused on surface solar radiation (SSR) (Dutton and Bodhaine, 2001; Wild, 2009) have evidenced a decreasing trend in solar radiation at Earth's surface after the mid-1950s followed by an increasing trend from the mid-1980s. These inter-decadal periods with prevailing reduction and enhancement of SSR are also known as solar dimming and brightening, respectively. These variations in SSR are not externally forced by variations of the sun's radiative output (Lockwood and Fröhlich, 2007), and they are therefore expected to be internal to the Earth's atmosphere. Ruckstuhl et al. (2008) suggest they are a consequence of changing atmospheric transmittance mainly driven by atmospheric aerosol content changes (natural or anthropogenic), cloud changes or a combination of both effects. In this context, there are a number of studies that highlight the existence of a peak in aerosol concentrations in 1988–1990 as a consequence of the increasing anthropogenic aerosol emissions during the solar dimming (Streets et al., 2009; Wild, 2009; Nyeki et al., 2012). This increase in aerosol content was partly responsible for the widespread reduction in sunlight at the Earth's surface observed in some regions, mainly in central Europe, Africa and some locations in Asia. Subsequently, the air quality regulations introduced since 1980s in many European countries caused an important reduction in atmospheric aerosol pollution and therefore the consequent brightening over Europe from the late 1980s onward. However, the cause of dimming/brightening is not fully understood and the large-scale significance of this phenomenon is called into question. Thus, the relative importance of aerosol and/or cloud effect on the dimming/brightening is probably different across the globe. There are some studies in the literature aiming to clarify it, such as those performed by Dutton and Bodhaine (2001), Ruckstuhl et al. (2008) and Nyeki et al. (2012). The last two studies used Aerosol Optical Depth (AOD) information at high altitude sites, but they did not find significant trends in this parameter in the periods 1995–2005 and 1995–2010, respectively. Longer aerosol concentration series are required to detect their effects on dimming/brightening processes at high altitude.

Finally, there are other global or regional trends in aerosol concentrations that could affect the overall record. They are prone to be related to the strong decrease in emissions observed in the developed countries caused by air quality regulations. However, the evolution of aerosols in the atmosphere result from highly non-linear mechanisms and thence the trend analysis from a long-term record is a complex task (Collaud Coen et al., 2013). Small trends might be detected as a consequence of changes in long-range transport of dust and pollution, changes in local pollution sources, increased emission of pollutants in developing countries or, more difficult to discern, long-term climate variability processes linked to aerosol transport.

At present, poor data quality and changes in the methodology of measurements of the existing databases are considered the principal problems to differentiate reliably significant aerosol trends from natural variability in aerosol concentration. Thus, long records of quality controlled aerosol loadings datasets are needed to better understand the causes of the observed trends. Due to the lack of availability of reliable multi-year observations, long-term trend analyses of aerosols concentration at present rarely exceed more than 20 years. There exists in the literature some exceptions of long and continuous aerosol records like the oldest AOD series extracted by Ohvril et al. (2009) in Russia, Ukraine and Estonia using measurements of direct solar radiation (Lindfors et al. , 2013) or the AOD series measured in Mauna Loa (Shaw , 1979; Holben et al., 2001) by means of sun-photometry, and Barrow (Bodhaine and Dutton , 1993), using a four-wavelength nephelometer, both since 1976. Unfortunately, as is stated by Lindfors et al. (2013), such data exist only for few selected stations. It means that the existing datasets are difficult to be used for most scientific assessments because they are fragmented or not long enough for these purposes.

Implementation of systematic measurements of aerosol properties at sites with regional or global representation began in the mid-1970s at several remote locations, such us South Pole, Mauna Loa or Barrow (Collaud Coen et al., 2013). Historically, the most ambitious attempt to monitor background aerosol optical depth levels was organized under the World Meteorological Organization (WMO) Background Atmospheric Pollution Monitoring Network (BAPMoN) program, which operated from 1972 to 1992 and served as a precursor of the current WMO Global Atmospheric Watch (GAW) network (Wehrli , 2008). However, the low precision and stability of the earliest hand-held photometers soon dissuaded the continuity of long term programs to detect AOD trends with such instruments (WMO, 1994).

Ground-based sites in background conditions, far away from anthropogenic sources, are important for studying spatial and temporal variability of atmospheric aerosol properties as well as climate relevant changes and trends. In this context, Izaña is a key site to detect aerosol trends, and interannual variability of dust transport associated to climate variability. Izaña is a remote high-altitude site representative of free troposphere conditions associated to low aerosol concentration values (Rodríguez et al., 2009; Basart et al., 2009; Cuevas et al., 2013). Due to its location, in the North Atlantic subtropical region, it is also suitable to study exchange processes between tropics and mid-latitudes and long term monitoring of frequency and intensity of Saharan dust outbreaks over the north Atlantic.

In this work we have recovered a 37 year series of monochromatic AOD from long-term solar irradiance measurements performed with the solar spectrophotometer Mark-I, continuously operated since 1976 at Izaña mountain. AOD is the simplest variable to remotely assess the aerosol loading in the atmosphere from ground-based instruments (Holben et al., 2001) because it represents the vertical integral of the aerosol direct irradiance extinction as a measure of atmospheric transmittance (Ruckstuhl et al., 2008). This series encompasses the period between 1976 and 2012. From 2001 onward accurate AOD measurements performed with various sunphotometers are available at the Izaña Atmospheric Observatory, such as those from GAW-Precision Filter Radiometer (PFR), since 2001, and from Cimel-AErosol RObotic NETwork (AERONET), since 2003. Thence, many comparative studies can be performed to accurately validate the recovered AOD dataset. Accordingly, we have studied in this paper the ability of the astronomical Mark-I spectrometer to determine AOD data comparable with AOD measured with sunphotometers of accepted aerosol monitoring networks. As a part of the validation, a full comparative analysis have been carried out between AOD quasi-simultaneously derived from Mark-I, AERONET and PFR.

2 The Izaña site: astronomical and atmospheric observations

Tenerife is one of the oldest atmospheric and astronomical monitoring sites worldwide. The atmospheric monitoring is carried out at the Izaña Atmospheric Observatory (IZO), from the Izaña Atmospheric Research Centre (IARC; http://izana.aemet.es), managed by the State Meteorological Agency of Spain (AEMET; http://www.aemet.es), meanwhile the SolarLab is a telescopic installation for solar observations owned by the Instituto de Astrofísica de Canarias (IAC; http://www.iac.es) at the Teide Observatory (OT). Both centers are located at a distance of 1.5 km from each other.

IZO, located at an average altitude of 2370 m above sea level, provides atmospheric measurements representative of free troposphere conditions due to the quasi-permanent subsidence regime typical of the subtropical region. It entails a frequent winds flow in the lower troposphere resulting in a strong and persistent temperature inversion layer, normally located between 800 and 1500 m a.s.l., below the Izaña's level. It prevents the pollution from the lower part of the island and separates a dry free troposphere from a relative fresh and humid oceanic boundary layer (Basart et al., 2009; Cuevas et al., 2013). Thus, Izaña is a key location to perform atmospheric studies with significant atmospheric measurements over the last 30 years under international programmes (i.e., WMO-GAW, NDACC -Network for the Detection of Atmospheric Composition Change-). This remote high-altitude site is indicative of the global aerosol transport because it is located within the broad "dust belt" that extends from the eastern subtropical Atlantic eastwards through the Saharan Desert to Arabia and Southwest Asia (Basart et al., 2009) and it is close to the subtropical barrier, allowing us to study exchange processes between the tropics and mid-latitudes. As Basart et al. (2009) found, aerosol background conditions at IARC are associated to low AOD values, $\approx 85\,\%$ under 0.15, meanwhile larger values are associated to dust events more likely to occur during summertime. Thus, a long-range dust transport above trade wind inversion layer is observed from early summer to early autumn.

Many ancillary data are collected at IARC for research purposes. Those of interest for this study are accurate AOD measurements from the GAW-PFR, starting in 2001, and from CIMEL/AERONET since 2003, with a short campaign during summer 1997.

SolarLab contains a total of six instruments which operate continuously on a daily basis, and in some cases for more than 35 years. The IAC's Helioseismology group is one of the pioneer groups in this scientific domain. They participated in 1979, jointly with colleagues of the University of Birmigham, in the key discovery that allowed the fast development of Helioseismology, the discovery of the global nature of the 5 min solar oscillations and therefore their identification as the "Sun's eigenmodes" (García et al., 2007). The first instrument ("Mark-I"), contributed to this find through the high precision measurements of the radial velocity of the Sun-as-a-star.

3 The Mark-I spectrophotometer

Mark-I is a reference instrument in helioseismology (Pallé and Roca-Cortés, 2012) specifically designed to study the small radial velocity fluctuations of the Sun's photosphere produced by solar oscillations and the measurements of the General Theory of Relativity on the Earth's surface. Mark-I is as a potassium-based resonance scattering spectrometer developed at Birmingham University and extensively described in Brooks et al. (1978), being currently the seed node of the Birmingham Solar Oscillations Network (BiSON) (Pallé and Roca-Cortés, 2012). Mark-I employs a magneto-optics filter to study the solar surface and the apparent Doppler velocity of the 769.9 nm resonance line of neutral potassium atom in the light integrated over the entire Sun (viewed as a star). It offers us long-term stability and very high precision in comparison to instruments based on interference filters. It has operated since 1976, with its main optical components remaining basically unchanged.

This is an equatorially mounted spectrophotometer in which sunlight is fed into the instrument using a coelostat (Fig. 1a), an arrangement of open air flat mirrors with changeable orientation during the day to follow the course of the sun. Following the diagram in Fig. 1b, the output beam is reflected, through a hole in the wall, into the spectrometer. Solar light passes through a 1.5 nm interference filter, centered at ≈ 770 nm. The light is then directed towards the spectrometer where it traverses a circular polarizer and an electro-optical light modulator (Pallé et al., 1992). The sunlight within this spectral range is affected mainly by atmospheric potassium and O_2 absorption. The absorption lines of these atmospheric constituents are displayed in Fig. 2a. It shows that the potassium line centered at 769.89 nm plays the most important effect in this spectral region, although O_2 absorption processes are also present. A stable vapor cell of potassium is placed in a longitudinal and permanent magnetic field (0.18 T), having a resonance line overlapping the solar absorption line (solid black curve in Fig. 2b). The magnetic field causes the two Zeeman components (blue and red curves in Fig. 2b) to sample the circularly polarized KI 7699 Å solar line. Mark-I measures alternatively each second the intensity of resonantly scattered light due to left-handed (L) and right-handed (R) circularly polarized incident light (Pallé and Roca-Cortés, 2012). Then, the ratio $r = \frac{L-R}{L+R}$ is calculated in blocks of 40 s, filtered by clouds and instrumental errors. Cloud screening process involved the detection and removal of cloud contaminated data by detection of variations in the photometric signal (R+L) and also in the solar radial velocity curve (R-L)/(R+L). It is carefully done manually day by day by the astronomer of Mark-I in order to ensure data quality. The ratio gives a measure of the lineshift corrected for any intensity fluctuation and thence it is linearly related to the velocity shift between the Sun and the laboratory. Since the narrow laboratory Zeeman components scans the steepest part of the broader solar line as Earth spins, the scattered light is also sensitive to the Sun-laboratory relative velocity.

At this paper purposes, the most important variables are measured by two photomultiplier tubes (Scattering PMT and transmission PMT on the diagram in Fig. 1b). The scattering PMT is placed in the transverse direction of the incident beam, taking the information of scattered light ($R+L$) at any direction as a consequence of the resonant scattering effect inside the vapor cell. The other one is set in the direction of the transmitted beam, taking the information of the transmitted light. We have scattered light measurements since 1976 but transmitted light information is limited to the period from 1984 to 2002. Since transmitted light is a strictly photometric magnitude and therefore directly related to the atmospheric extinction, we will preferably use it to recover AOD information. The information extracted from the scattered component has also a residual velocity contribution that could affect the accuracy of the retrieved AOD. However, due to the lack of transmitted component information for years 1976, 1977, 1980–1983 and 2003–2012, AOD was determined from scattered light in the remained period. Since this component is the sum of two monochromatic (R+L) measurements in the profile of the solar absorption line, it can also be used for AOD determination. As a result, we have a total of 5,786,031 solar measurements available to determine AOD information, which correspond to 8,084 days of measurements.

4 Calibration and methodology of AOD determination from Mark-I observations

Mark-I calibration was performed using the Langley technique (Shaw , 1983; Schmid et al., 1997; Holben et al., 1998). It is the most common approach in photometry to determine the extraterrestrial voltage $V_{0,\lambda}$ from monochromatic photometric measurements performed at different solar elevation angles or air masses. This method is based on the Beer–Lambert–Bouger Law which implies that the zero-air-mass photometer voltage ($V_{0,\lambda}$) can be determined by means of a plot analysis of these monochromatic measurements against air masses, provided the atmospheric turbidity remained constant and preferably low over the measurement's period.

Corrections for Rayleigh scattering (Kasten and Young, 1989; Bodhaine et al., 1999) and for mixed gases have been included in the calibration procedure.

We have performed such analysis for the transmitted component by selecting those days with AOD < 0.04 and correlation coefficients of the fitting plot ≥ 0.99. The only exception for these thresholds was set during the period from 1992 to 1993, when the Mt. Pinatubo eruption (June 1991) released huge amounts of volcanic aerosols into the stratosphere leading to an important decrease in solar radiation input as well as important positive anomalies in AOD. For this reason, the threshold in AOD for the Langley analysis was set in 0.3, an order of magnitude higher. Due to the high dispersion of extraterrestrial voltages V_0 obtained from the scattered component as a consequence of the Sun-laboratory velocity effect on this component, we have set the AOD threshold to perform a Langley using this component in 0.3, in order to have a higher number of daily calibration values to follow the variation in the instrument's calibration. In following sections we will discuss the suitability of those Langleys performed under such high aerosol content conditions.

The yearly V_0 variation of Mark-I solar spectrometer determined from the Langley analysis is shown in Fig. 3. A total of 2,462 V_0 s have been obtained using this technique, being the rest of days without V_0 information (about 70 % of them) recovered by means of a cubic spline smoothing process. These values were subsequently reprocessed when a deficient calibration was observed, being the most important calibration problems associated to the existence of a fictitious diurnal cycle on AOD data. This problem in calibration procedure was identified by Cachorro et al. (2004, 2008). Following these authors, we have considered a refinement procedure in order to improve the V_0 accuracy. In a first stage, we selected those days affected by a fictitious diurnal cycle on AOD, both with convex or concave behavior, characterized by an amplitude in AOD > 0.3. If a set of requirements are fulfilled (air mass $m < 7$, a minimum of 10 points to perform the fit after outliers removal, and correlation coefficient $r > 0.98$) a new calibration constant V_0' can be retrieved as the slope of the AOD vs. m^{-1} fitting plot. A second reprocessing step was included for AOD amplitude < 0.3 and AOD < 0 events. In this situation, V_0'' was determined considering similar requirements for m range, number of points and outliers than in the first stage, but less restrictive correlation coefficients ($r > 0.9$). Figure 3 clearly shows the important variation in the extraterrestrial voltage recovered from this astronomical instrument. It ranges from 3332 to 2.54×10^5. These significant variations in V_0 are directly associated with maintenance operations in the coelostat. It is worth mentioning transmitted component was used for calibration between 1984 and 2003, and scattered component in the remaining period. It is therefore expected changes in V_0 as a result of this change (see the variation between 1983 and 1984 as well as between 2003 and 2004). The most important change is observed in 1984, being attributed to the change in the scattering PMT gains made this year. In May 2011 the PMT for the scattered component was replaced, and then the consequent variation in V_0 is observed. Minor changes ($\sim 50\%$) are observed through the whole period, mostly attributed to the mirrors replacement and cleaning as well as the realignment processes. This appreciable variability in V_0 supposed a challenge to use photometric information from Mark-I to obtain a long-term series of AOD useful for climatic studies. As we will present in Sect. 5, we have used efficient techniques published in the literature to overcome this problem. Furthermore, these type of variations are frequent in the instruments involved in sunphotometry. In fact, the evolution of the V_0 at Izaña extracted from AERONET masters (from 2004 to 2014) presents variations in calibration constant up to $\sim 80\%$ as a result of the frequent replacement of AERONET "master" instruments at the Observatory.

5 Results

5.1 AOD validation

In order to compare AOD at $769.9\,\mathrm{nm}$ from Mark-I spectrophotometer with PFR and Cimel sunphotometers which take measurements at different wavelengths, we used the Ångström exponent following the Eq. (1). It relates the AOD value measured a two different wavelengths, λ and λ_0.

$$\mathrm{AOD}(\lambda) = \mathrm{AOD}(\lambda_0) \cdot \left(\frac{\lambda}{\lambda_o}\right)^{-\alpha} \tag{1}$$

AOD comparison using quasi-simultaneous data extracted from Mark-I, PFR and AERONET was performed to assess the ability of the solar spectrometer Mark-I to retrieve accurate long-term AOD information. We have considered coincident measurements performed with these instruments before 2013, entailing AERONET measurements in June and July 1997, and from 2003 to 2012, as well as PFR measurements in the period 2001–2012, both performed at IARC. Since PFR provides hourly-AOD data, we have considered quasi-simultaneous data when PFR and Mark-I measurements fall within $\pm 20\,\mathrm{min}$ range. It entails a maximum of 65 Mark-I observations into the $\pm 20\,\mathrm{min}$ window. A lower temporal range of $\pm 10\,\mathrm{min}$ was considered to compare AERONET/Mark-I data. It entails a maximum of 22 Mark-I measurements within this temporal window. A definition of the main statistics used in this paper is provided in Table 1. The scatterplots of daily AOD values from Mark-I, Cimel AERONET and PFR for each year (1997 and 2001-2012) are displayed in Fig. 4. The main skill scores of the validations are shown in Table 2. A low mean bias (MB) value of -0.008 was obtained for the whole period analysis in case of AERONET/Mark-I comparison, with a Pearson correlation coefficient (r) of 0.92. A total of 95,297 coincident measurements were used in this comparison. The annual comparison shows MB values ranging from -0.018 to 0.011 as well as high correlation coefficients ($r > 0.90$) and RSMEs below 0.030. The only exception was obtained for 2003, when a high RMSE value of 0.053 and a value of $r = 0.89$ were retrieved. This unexpected poor performance could be attributed to higher calibration inaccuracies of Mark-I data during this period.

Regarding the PFR/Mark-I comparison, a total of 14,260 matching cases were used, and a MB of 0.004 was obtained. A fairly good agreement is found between both instruments (Table 2), obtaining MB values ranging from 0.018 to -0.008. Correlation coefficients are similar to those retrieved in the AERONET/Mark-I comparison, with values > 0.83, with the exception of 2003, with a markedly lower r value ($r = 0.74$). These discrepancies with PFR data confirm the existence of calibration inaccuracies on Mark-I series during 2003 also detected in the AERONET/Mark-I comparison. The most important MB deviations are observed for 2011 with a value of 0.018. The high MB observed for 2008 was attributed to the sparsity of data. The skill scores in the rest of years revealed a MB ≤ 0.01.

In general, AOD from these instruments correlate quite well each other, with AOD differences between AERONET and Mark-I and between PFR and Mark-I lying within the range 0.01–0.02 (see Table 2). These differences are similar to those obtained between PFR and AERONET (0.01–0.02) reported by Barreto et al. (2013) and Kazadzis et al. (2014) and higher than those obtained by Nyeki et al. (2012). Taking into account these results we can be confident on the ability of Mark-I solar spectrometer to obtain AOD with relatively small differences in comparison with reference instruments.

5.2 Aerosol content impact on instrument's calibration

In order to assess the suitability of including days with high aerosol loads to perform Langley calibrations we have compared quasi-simultaneous and coincident AOD information determined from Mark-I observations (transmitted component) and PFR during 2002. As in the previous section, we have considered quasi-simultaneous data when PFR and Mark-I measurements fall within a temporal range of $\pm 20\,\mathrm{min}$. A total of 818 coincident AODs have been included which correspond to 92 different days. The MBs ($\mathrm{AOD_{PFR}}$ vs. $\mathrm{AOD_{Mark\text{-}I}}$) obtained in low aerosol content events (0.008 for AOD ≤ 0.04) is of the same order of magnitude to those obtained in case of higher turbidity conditions (-0.005 for $0.04 <$ AOD < 0.3) (Table 3). In principal, the V_0 values obtained in days affected by rather high aerosol loads could be considered as not suitable, and would result in wrong AOD values. However, our results show that calibration errors are not dependent on the aerosol load, provided aerosol concentration remained constant. Despite aerosol stability conditions are most likely met under low AOD cases, according to our experience they can be also reachable under relatively high aerosols (mineral dust) content. This stability condition is assured by restricting the calibration analysis to highly correlated Langley calibrations ($r \geq 0.99$). This is an outstanding result since it tell us we can obtain reliable V_0 in long periods of relatively high AOD (up to 0.3), as the recorded after the Mt. Pinatubo eruption affecting the whole year 1992.

6 AOD series from MARK-I

Data consistency relies on in its homogeneity, assuring that data variability is not caused by changes in the instrument. The 37 year AOD series determined with Mark-I as well as the long term monthly variation of the homogenized data set are shown in Fig. 5. The last one was calculated using the methodology proposed by Lanzante (1996). It is an iterative procedure that makes possible to detect outliers and inhomogeneities in the median, evaluate the signal to noise ratio, and finally eliminate those inhomogeneities with a confidence level of 99 %. We can see in Fig. 5 the impact of the two most important volcanic eruptions: El Chichón (1982) and Mt. Pinatubo (1991).

Although an accurate analysis of the long term series is out of the scope of this work, we have made a preliminary AOD analysis as well as a rough assessment of AOD trends from Mark-I data. Since IZO is strongly affected by Saharan dust intrusions during spring and summertime, being these events driven by atmospheric processes with strong interannual variability, we consider more appropriate to include in this trend analysis only Mark-I information corresponding to wintertime. This season (from December to February) is virtually unaffected by dust conditions. The trend analysis was performed for the period 1984-2012 following Ruckstuhl et al. (2008), who estimated the trends for different time periods by a fitting plot of monthly mean AOD with a Least Mean Square approximation. This methodology for trend detection can be summarized in the Eq. (2), where Y_t represents the climate variable under study (AOD) at time t, μ is a constant term, ω is the linear AOD trend, S is the seasonal term and N_t is the monthly mean noise of the time series, that is assumed to be autoregressive of the order of the model. Further details of this methodology can be found in Weatherhead et al. (1998). We have not included in the trend analysis data from 1991 to 1994 because of the Mt. Pinatubo eruption, occurred in 1991. In such volcanic events the effect of the eruption decays with time (Weatherhead et al., 1998) and therefore introduces non-linear terms not considered in the linear model given in Eq. (2).

$$Y_t = \mu + \omega \cdot X_t + S + N_t, \qquad t = 1, \ldots, T \tag{2}$$

We have estimated the decadal AOD median at Izaña, presented in Table 4. AOD remained nearly stable at 0.02 since 1984, with values of 0.022 in 1984–1993 decade, 0.024 in 1994–2003 decade and 0.027 in the period 2004–2012. Our results do not show the expected decrease in AOD as a result of the solar brightening effect, as stated, for example, by Streets et al. (2009) who observed a reduction in aerosol concentration over Europe at 550 nm from 0.31 in 1980 to a stable value of 0.26 in 2000–2006. However, we have to keep in mind, first, that IZO is representative of subtropical free troposphere conditions, with associated very low AOD background values, and hence less affected by continental pollution, and secondly, the decisive role played by Saharan dust intrusions in our region, basically in summertime, which shows significant inter-annual and decadal variations modulating the long-term AOD series.

The effect of the Mt. Pinatubo eruption is clearly detected in the AOD record of Fig. 5. El Chichón eruption signal on AOD record is not as apparent as the previous one because of the gaps in the data series. From the homogenized monthly series we have assessed the AOD anomalies caused by these two important volcanic events. These anomalies are the AOD differences regarding the average AOD of the period 1984–1993. The AOD anomaly observed after El Chichón eruption in 1982 was 0.023. No wintertime data was available to evaluate anomalies in 1983. In the case of Mt. Pinatubo, the anomalies observed after the eruption were more important and persistent than for El Chichón eruption, recording a peak impact in 1992 with an AOD anomaly of 0.049 ($\sim$ 120% far from background conditions). The effects of Mt. Pinatubo eruption in AOD was not significantly reduced until early 1993. The highest monthly AOD value observed at Izaña as a result of the Pinatubo eruption was measured in July 1992 (0.14). These values are consistent with AOD measurements at 500 nm at Mauna Loa (Hawai) reported by Holben et al. (2001). These authors present peak AOD values at 500 nm of 0.19 and anomalies in AOD $\sim$ 0.1 (Dutton et al. , 1994).

Regarding the trend analysis in whole period, our results indicate there is no significant trend in the AOD at IZO from 1984 to 2012. This result is consistent with the negligible and no statistically significant AOD trend found by Ruckstuhl et al. (2008) by using long-term aerosol concentration measurements at three high-altitude stations. Nyeki et al. (2012) did not observe either a significant decrease in aerosol concentration in the period 1995–2010 at the same three high-altitude stations after a new re-calibration procedure.

These results will be subject of detailed analysis and assessments in future studies, incorporating more information of in-situ aerosols at IZO, backward trajectories and other ancillary information accounting for Saharan dust intrusions over the Canary Islands since these modulate the AOD variations at IZO.

7 Summary and conclusions

In this work we have assessed the feasibility to determine long term series of AOD from long-term irradiance measurements using an astronomical spectrometer. This astronomical device, named Mark-I, was specifically designed as a reference instrument in helioseismology. It accounts for a magneto-optics filter and operates using the method of resonant scattering, based on atomic transitions of sunlight incident into a potassium cell. It offers long-term stability and very high precision in comparison to instruments based on interference filters. However, the use of an exposed mirror arrangement to collect the sunlight introduces important changes in the instrument calibration that must be corrected. In addition, due to the Sun-laboratory velocity effect on the scattered component, high dispersion in V_0's introduced additional difficulties in the Mark-I calibration. To solve these problems, we have proposed a quasi-continuous Langley calibration procedure which, in principle, is not a standard procedure because it entails calibration under relatively high AOD conditions. However, our results indicated that calibration errors are not dependent on the aerosol load and therefore, V_0 can be calculated using this technique in those days with relatively high turbidity (AOD up to 0.3), provided aerosol concentration remains constant. This result is important in order to extend and assure calibrations for relatively long periods of time when it is not possible to find days with AOD < 0.05 suitable to perform Langley calibrations, such as after volcanic aerosols released into the atmosphere associated to important volcanic eruptions with global impact similar to Mt. Pinatubo, with lasted from 1991 to 1993. In addition, we have considered a refined procedure to improve the calibration which corrects for deficient calibration as well as fictitious diurnal cycle on AOD data.

AOD determined from Mark-I spectrometer using these procedures has been compared with quasi-simultaneous AOD measurements using reference instruments (Cimel-AERONET and PFR). The 11 year and 12 year comparison between AERONET and Mark-I and between PFR and Mark-I, respectively, showed MB values ≤ 0.01 in both cases, what constitutes an excellent result. The yearly validation performed in all cases by means of Cimel-AERONET/Mark-I and PFR/Mark-I comparison showed low MB values ≤ 0.02, within the precision of both sunphotometers (Cimel and PFR) and high correlation coefficients (generally $r > 0.90$). The exception is 2003, when some calibration problems in Mark-I, that affected the AOD extracted using both the transmitted and the scattered component, were detected. This validation procedure shows the ability of Mark-I to retrieve an AOD series which presents small differences in comparison with those of reference instruments (PFR and Cimel). This allowed us to extend the long-term AOD series at Izaña site with PFR and Cimel (since 2001 and 2003, respectively) back until 1976 using AOD data from Mark-I. This is a major achievement to carry out future studies of inter-annual and decadal variations of AOD.

We have performed a preliminary trend analysis for IZO using wintertime Mark-I data. We have obtained an AOD nearly stable of 0.022 since 1984, as well as a negligible trend in AOD in the time period from 1984 to 2012. The decadal trend analysis performed using Mark-I data revealed that no significant trend in AOD was observed at IZO. We have also estimated the impact of the major volcanic events occurred since 1976: the eruptions of El Chichón (1982) and Mt. Pinatubo (1991). We observed AOD anomalies due to El Chichón eruption up to 0.023. More important and persistent anomalies up to 0.049 were associated to the eruption of Mt. Pinatubo.

Further analysis, incorporating information of dust events, is required in order to ensure the existence of a statistically significant decrease in AOD since 1980s as a confirmation of the solar brightening in our region, as well as other statistically important trends on AOD. This is especially critical in the subtropical region over the North Atlantic since Saharan dust intrusions undergo large inter-annual and decadal variations modulating AOD variations.

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Table 1. Definitions of the statistics used in this study. f_i and Obs_i represent the variable under validation and the reference value, respectively.

Score	Statistics Equation	Range	Perfect score
Mean Bias (MB)	$\mathrm{MB} = \frac{1}{N}\sum(f_i - \mathrm{Obs}_i)$	$-\infty$ to $+\infty$	0
Root mean square error (RMSE)	$\mathrm{RMSE} = \sqrt{\frac{1}{N}\cdot(f_i - \mathrm{Obs}_i)^2}$	0 to ∞	0
Pearson correlation coefficient (r)	$r = \frac{\sum(f_i - \overline{f_i})\cdot(\mathrm{Obs}_i - \overline{\mathrm{Obs}_i})}{\sigma_{f_i}\cdot\sigma_{\mathrm{Obs}_i}}$	-1 to 1	1

Table 2. Skill scores from AERONET/PFR versus Mark-I comparison (transmitted component for 1997, 2001 and 2002, and scattered component from 2003 to 2012): Mean bias (MB), RMSE, Pearson correlation coefficient (r) and number of cases (n). Total row indicates the scores obtained using the whole period.

Instrument	Year	Mark-I Spectrometer MB	RMSE	r	n
AERONET Cimel	1997	0.006	0.021	0.91	1973
	2003	0.011	0.053	0.89	11 743
	2004	−0.014	0.026	0.95	3989
	2005	−0.013	0.023	0.96	6521
	2006	−0.012	0.022	0.96	7587
	2007	−0.011	0.021	0.94	12 304
	2008	−0.008	0.025	0.94	14 947
	2009	−0.018	0.030	0.90	9521
	2010	−0.015	0.020	0.97	8321
	2011	−0.009	0.020	0.96	9793
	2012	−0.007	0.021	0.96	8598
	TOTAL	−0.008	0.034	0.92	95 297
PFR	2001	0.001	0.014	0.98	376
	2002	0.004	0.020	0.92	1597
	2003	−0.008	0.019	0.74	550
	2004	0.001	0.032	0.86	886
	2005	0.007	0.029	0.94	565
	2006	−0.003	0.018	0.97	1638
	2007	0.008	0.023	0.93	1942
	2008	0.013	0.022	0.83	282
	2009	0.006	0.027	0.91	1281
	2010	0.002	0.015	0.97	1695
	2011	0.018	0.024	0.95	1975
	2012	0.004	0.018	0.96	1473
	TOTAL	0.004	0.022	0.94	14 260

Table 3. Bias and cases number of the daily-coincident comparison between PFR and Mark-I (transmitted component) AOD extracted during 2002.

AOD range	MB	#cases
≤ 0.04	0.008	635
$(0.04, 0.1]$	−0.005	75
$(0.1, 0.2]$	−0.005	70
$(0.2, 0.3]$	−0.005	38

Table 4. Median and standard deviation (σ) of monthly-mean homogenized AOD data set during wintertime (December, January and February).

Decade	Median AOD	$\boldsymbol{\sigma}$
1984–1993	0.022	0.012
1994–2003	0.024	0.014
2004–2012	0.027	0.014

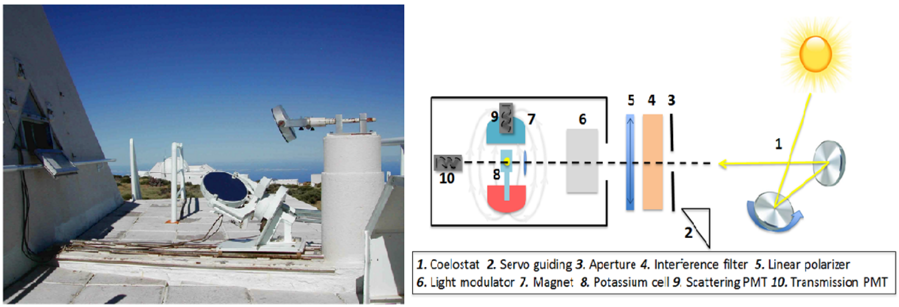

Figure 1. Mark-I coelostat system at the entrance of the spectrometer, on the left, and short diagram of the main constituents of the Mark-I spectrometer on the right.

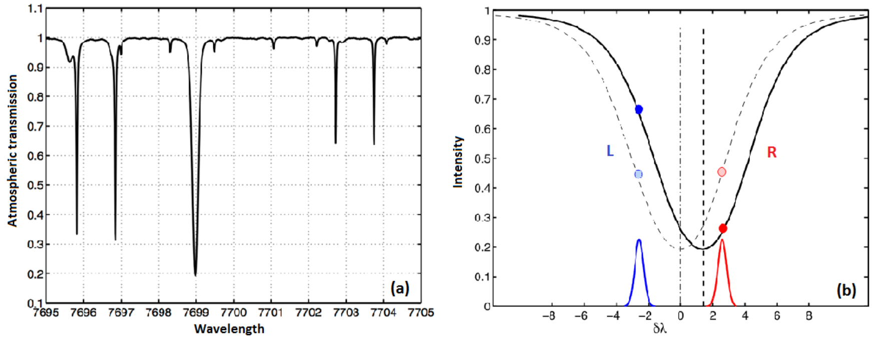

Figure 2. (a) Atmospheric transmission within the Mark-I interference filter bandwidth, and **(b)** the relative displacement of solar line (solid black curve) with respect to the laboratory line (dashed curve) measured as a function of R and L.

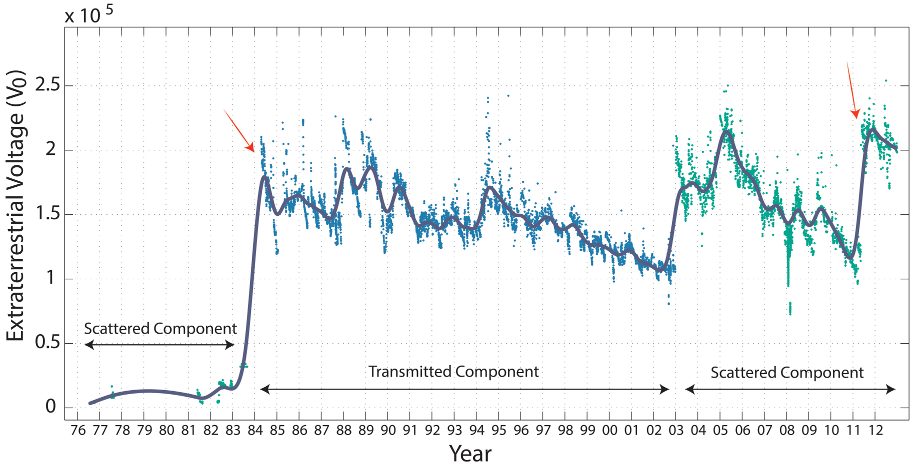

Figure 3. Mark-I extraterrestrial voltage (V_0) variation extracted from the scattered (in green) and the transmitted component (in blue) within the period 1976–2012. The smoothing cubic spline is displayed with the violet line. Red arrows mark a change in the PMT of Mark-I.

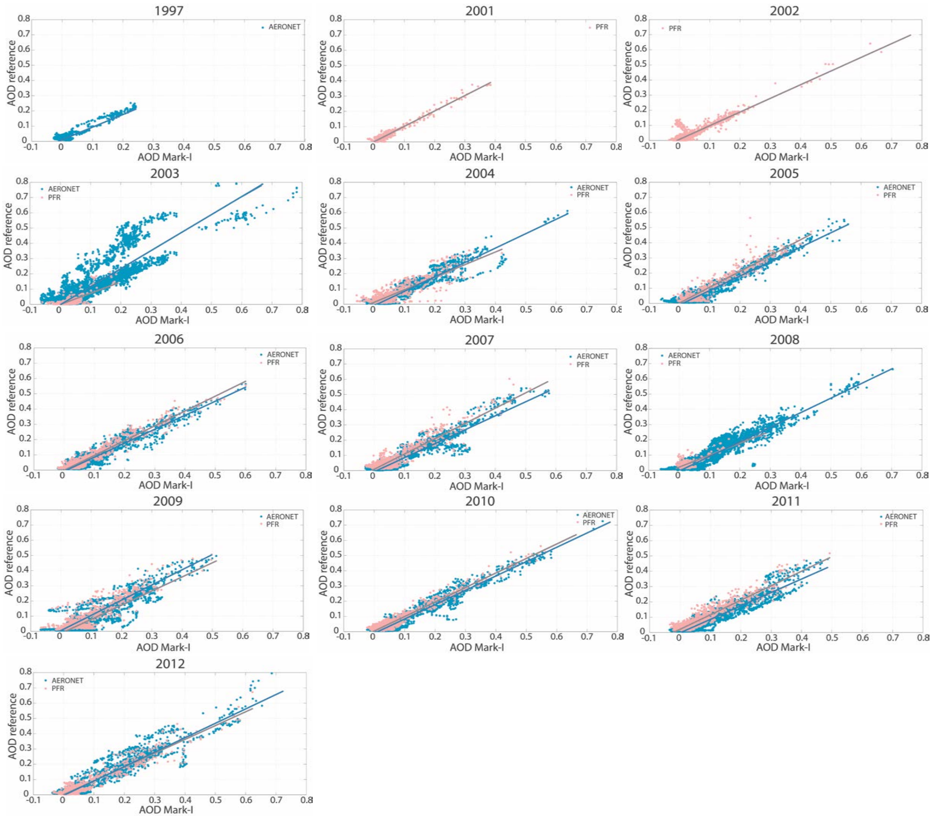

Figure 4. Scatterplots of daily AOD values from Mark-I against Cimel AERONET (blue) and PFR (pink) for each year (1997 and 2001-2012).

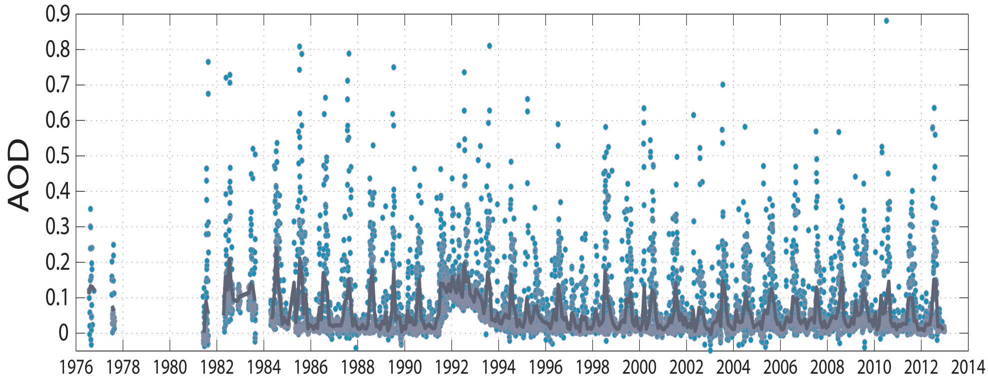

Figure 5. Long-term daily mean AOD (at 769.9 nm) data series at Izaña extracted from Mark-I scattered data (from 1976 to 1983, as well as from 2003 to 2012) and transmitted data (from 1984 to 2002). Monthly mean of the homogenized AOD series is displayed with the violet line. No available data from 1978 to 1980.