Aerosol radiative effects in the ultraviolet, visible, and near-infrared spectral ranges using long-term aerosol data series over the Iberian Peninsula

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Abstract

A better understanding of the aerosol radiative properties is a crucial challenge for climate change studies. This study aims to provide a complete characterization of aerosol radiative effects in different spectral ranges within the shortwave (SW) solar spectrum. For this purpose, long-term datasets of aerosol properties from six AERONET stations located in the Iberian Peninsula (Southwestern Europe) are analyzed in term of climatological characterization and trends. Aerosol information is used as input to the libRadtran model in order to determine the aerosol radiative effect at the surface in the ultraviolet (ARE_UV), visible (ARE_VIS), near-infrared (ARE_NIR), and the entire SW range (ARE_SW) under cloud-free conditions. Over the whole Iberian Peninsula, yearly aerosol radiative effects in the different spectral ranges are: $-1.1 < \text{ARE}_{\text{UV}} < -0.7 \text{ W m}^{-2}$, $-5.7 < \text{ARE}_{\text{VIS}} < -3.5 \text{ W m}^{-2}$, $-2.6 < \text{ARE}_{\text{NIR}} < -1.6 \text{ W m}^{-2}$, and $-8.8 < \text{ARE}_{\text{SW}} < -5.7 \text{ W m}^{-2}$. The four variables showed positive statistically significant trends between 2004 and 2012, e.g., $\text{ARE}_{\text{SW}}$ increased $+3.6 \text{ W m}^{-2}$ per decade. This fact is linked to the decrease in the aerosol load, which presents a trend of $-0.04$ unit of aerosol optical depth at 500 nm per decade, hence a reduction of aerosol effect on solar radiation at the surface is seen. Monthly means of ARE show a seasonal pattern with larger values in spring and summer. The aerosol forcing efficiency (AFE), ARE per unit of aerosol optical depth, is also evaluated in the four spectral ranges. AFE exhibits a dependence on single scattering albedo and a weaker one on Ångström exponent. AFE is larger (in absolute value) for small and absorbing particles. The contributions of the UV, VIS, and NIR ranges to the SW efficiency vary with the aerosol types. Aerosol size determines the fractions of $\text{AFE}_{\text{VIS}}/\text{AFE}_{\text{SW}}$ and $\text{AFE}_{\text{NIR}}/\text{AFE}_{\text{SW}}$. VIS range is the dominant region for all types, although non-absorbing large particles cause a more equal contribution of VIS and NIR intervals. The $\text{AFE}_{\text{UV}}/\text{AFE}_{\text{SW}}$ ratio shows a higher contribution for absorbing fine particles.
1. Introduction

Atmospheric aerosol particles can absorb and scatter part of the total amount of solar radiation entering the Earth's atmosphere. In fact, aerosols directly influence the Earth's energy budget and act as cloud condensation nuclei modifying the cloud structure (e.g., Forster et al., 2007). Aerosols can either be produced by ejection into the atmosphere or by physical and chemical processes within the atmosphere. Aerosol particles affect the radiative field by attenuating the direct component thereby enhancing (or reducing under a highly absorbing aerosol) the diffuse one. They also produce indirect effects (AIE) by perturbing the Earth's atmospheric radiative balance by modulating cloud albedo and fraction. The AIE can be viewed as a series of processes linking various variables such as aerosol mass, cloud condensation nuclei concentration, ice nuclei concentration, water phase partitioning, cloud optical properties, etc. (Penner et al., 2001).

The aerosol radiative effect (ARE) is defined as the change in net radiation due to changes in atmospheric aerosol properties and content. This is a key quantity in the determination of climate change (e.g., Hansen et al., 1998). Most studies dealing with ARE have focused on discrete wavelengths, whole shortwave (SW) solar radiation spectrum (e.g., Rajeev and Ramanathan, 2001; García et al., 2008; di Sarra et al., 2008; Cachorro et al., 2008; Foyo et al., 2014), longwave (LW) radiation (e.g., Panicker et al., 2008; di Sarra et al., 2011; Antón et al., 2014), ultraviolet (UV) interval (e.g., Hatzianastassiou et al., 2004; Kazadzis et al., 2009; Nikitidou et al., 2013), and visible (VIS) range (e.g., Jayaraman et al., 1998; Horvath et al., 2002; Bush and Valero, 2003; Meloni et al., 2003). With regards to surface SW radiative effect (ARE_{SW}), Di Biagio et al. (2010) obtained the maximum radiative daily effects for different aerosol types in the Central Mediterranean in the period 2004-2007: -61 Wm^{-2} (desert dust aerosols), -26 Wm^{-2} (urban/industrial - biomass burning aerosols) and -43 Wm^{-2} (mixed aerosols). All these negative figures point out a cooling of the Earth's surface. Aerosol radiative effects in the LW (ARE_{LW}) are expected to be smaller than in the
SW and with positive sign (see, e.g., di Sarra et al., 2011; Antón et al., 2014). Hence, this heating effect at the surface can partly offset the cooling induced in the SW range. With respect to the ARE for the UV range (ARE$_{\text{UV}}$), Nikitidou et al. (2013) analyzed the ARE in two different spectral regions in the UV range, 300-315 and 315-360 nm. They found a stronger attenuation in the UV-B than in the UV-A, however being both solar zenith angle (SZA) dependent because of different photon paths.

The main goal of this study is to evaluate the ARE at the surface over the Iberian Peninsula, which is a region of great interest because of its geographical position in Southwestern Europe, near the African continent and the interface between the Atlantic Ocean and the Mediterranean Basin. Thus, it is affected by frequent desert dust intrusions which modulate their aerosol climatology (Toledano et al., 2007a, Bennouna et al., 2013; Pey et al., 2013; Valenzuela et al., 2012). In addition, this area is also affected by a great variety of air masses loaded with different aerosol types: clean continental, polluted plumes of central Europe, and marine aerosols. Hence, aerosol climatology at six stations (Palencia, Barcelona, Cabo da Roca, Évora, Granada, and El Arenosillo) is also carried out for different time periods between 2001 and 2012. Aerosol radiative effects as well as their efficiency are calculated in four regions of the solar spectrum (ultraviolet, visible, near-infrared, and shortwave) and the relative contribution of each range with respect to the whole solar spectrum is analyzed as a function of the aerosol properties. Therefore, this study is intended to contribute to the understanding of the aerosol impact on radiative budget over the Iberian Peninsula.

This article presents the following outline: detailed descriptions of the aerosol stations and the database used are performed in Section 2; Section 3 includes the followed methodology; the results obtained in the different analyses about the climatology of aerosol properties, aerosol radiative effects, and aerosol forcing efficiencies are shown and discussed in Sections 4, 5, and 6, respectively. Finally, the main conclusions of this article are summarized in Section 7.
2. Columnar aerosol optical data

The aerosol data are obtained from the Aerosol Robotic Network (AERONET) (Holben et al., 1998). Six AERONET sites operating in the Iberian Peninsula are selected in this study: Palencia, Barcelona, Évora, Cabo da Roca, Granada and El Arenosillo (see Table 1), all of them with a minimum of 8 years of data sets of continuous observations. These sites present the largest records of aerosol properties in the Iberian Peninsula in the AERONET network.

The standard instrument used in AERONET is the Cimel CE-318 radiometer. It performs direct sun measurements at several wavelengths in the spectral range 340-1020 nm. Furthermore, the instrument also measures sky radiance in the solar almucantar and principal plane configurations at 440, 670, 870 and 1020 nm wavelengths. A detailed description of this instrument was provided by Holben et al. (1998). The direct sun observations are used to derive the spectral aerosol optical depth (AOD) and the corresponding Ångström exponent. The sky radiances together with the AOD are employed to retrieve a set of aerosol optical and microphysical properties via inversion methods (Dubovik and King, 2000; Dubovik et al., 2006). These include particle size distribution, complex refractive index, single scattering albedo (SSA), phase function, asymmetry parameter, fraction of non-spherical particles, etc. (see http://aeronet.gsfc.nasa.gov/new_web/Documents/Inversion_products_V2.pdf). Data are provided in three database levels: 1.0 (raw data), 1.5 (cloud-screened) and 2.0 (cloud-screened and quality assured).

The calibration of these instruments is performed following AERONET protocols by AERONET-NASA, PHOTONS and RIMA networks every 12 months of operation approximately. The estimated uncertainty is 0.01-0.02 for AOD (larger at shorter wavelengths) and ~5% for the sky...
radiances (Holben et al., 1998). The SSA has an absolute uncertainty about 0.03-0.07 depending on
the aerosol load and type (Dubovik et al., 2000).

Level 2.0 aerosol optical depth data have been used in this work. However, it is well-known that
when level 2.0 inversion data are used, the number of available observations of single scattering
albedo (SSA) and asymmetry factor (g) is quite limited because these variables are only considered
reliable when AOD_{440nm} > 0.4 \(^1\). Such AOD is mainly reached in the study region during Saharan
dust or biomass burning events, therefore we would not have information on SSA and g for other
conditions. To solve this issue, we have reduced the threshold of the level 2.0 inversion products.
For this, we started with the level 1.5 data (for those quality-assured almucantar data that reached
level 2.0) and applied the same criteria used by AERONET to elaborate the level 2.0 regarding the
number of symmetrical angles, retrieval error and solar zenith angle (see

However, a less restrictive threshold is applied to the AOD, which we restricted to cases with
AOD_{440nm} > 0.15, instead of 0.4. This choice must be considered a compromise between the amount
and the quality of the data. This kind of approach has been adopted by other authors using
AERONET absorption data (e.g. Mallet et al., 2013). The threshold of 0.15 seems adequate
analyzing the typical values of the AOD in the Iberian Peninsula (e.g., Bennouna et al., 2011;
Obregón et al., 2012), because it can be considered a value to separate background aerosol
conditions from episodic events with moderate or high aerosol loadings. The level 1.5-filtered data
of SSA and g are daily averaged in order to have one value per day. In these conditions, the
estimated uncertainty of the single scattering albedo is ±0.05-0.07 (Dubovik et al., 2000).

Furthermore, for those days presenting level 2.0 data but also measurements in the 1.5-filtered level,
we tested the uncertainty of our approach. We evaluated the difference in the SSA values of the

\(^{1}\) Other inversion products, like the volume size distributions, are provided for all AOD levels.
level 1.5-filtered data with respect to the closest level 2.0 data. The mean relative differences in the SSA values between both methodologies are smaller than 1%, being in the same order that the inversion uncertainty.

Lastly, when the AOD is low (<0.15 at 440 nm), there is no reliable information on the absorption properties in the almucantar retrievals. Such low AOD is typical in our study region (e.g. almost 70% of observations at Palencia, Granada and Évora are below this threshold). If only cases with AOD$_{440nm}>0.15$ are considered in our study, the derived aerosol radiative effect would be unrealistically large. To overcome this problem of representativeness, fixed values of SSA (0.90) and g (0.75) have been used for the cases with AOD < 0.15 at 440 nm, considering typical values for continental, desert, and maritime aerosols (e.g., Hess et al., 1998). In spite of the associated uncertainties, our approximation (daily level 1.5-filtered values of these aerosol properties for AOD > 0.15 together with a typical fixed value for low AOD cases) provides a good characterization of the aerosol absorption of the particles present in the atmosphere. The data products and AERONET database level are summarized in Table 2, where the estimated absolute uncertainties of AOD and SSA are also provided.

3. Methodology

The ARE calculations are performed in the ultraviolet (ARE$_{UV}$, 280-400 nm), visible (ARE$_{VIS}$, 400-700 nm), near-infrared (ARE$_{NIR}$, 700-2800 nm), and shortwave (ARE$_{SW}$, 280-2800 nm) intervals. For this purpose, cloud-free simulations are carried out by means of a radiative transfer code.

The libRadtran model (Mayer and Kylling, 2005) has been shown to be a useful tool for obtaining solar radiation data, presenting high accuracy in both cloudy and overcast conditions (e.g., Mateos et al., 2013b, 2014; Román et al., 2014). Version 1.7 of the libRadtran is used in this study with
inputs of aerosol, total ozone column (TOC), precipitable water vapor column (PWC), and surface albedo data. We performed simulations of ultraviolet (280-400 nm), visible (400-700 nm), near-infrared (700-2800 nm), and shortwave (280-2800 nm) radiation during the periods indicated in Table 1. Total ozone column is provided by the Ozone Monitoring Instrument (OMI) and Total Ozone Mapping Spectrometer (TOMS). Daily values of these instruments are obtained from the Daily Level 3 Global Gridded products, which are downloaded using the Giovanni application (http://disc.sci.gsfc.nasa.gov/giovanni). Level 2.0 AERONET PWC data are used in the calculations. The uncertainty of this parameter is 10-15% (Holben et al., 1998). In addition, retrievals of surface albedo at 440, 675, 870 and 1020 nm from the AERONET algorithm are also used in this work. For land surface cover, this algorithm relies on the Lie–Ross model (Lucht and Roujean, 2000), but considering the bidirectional reflectance distributions from MODIS (Moody et al., 2005).

Aerosol properties obtained from AERONET measurements are also used as input to the libRadtran model. Ångström coefficients, α and β, are utilized to compute a spectral aerosol optical depth in the wavelengths of interest (Schuster et al., 2006). Ångström exponent α is obtained with the measurements between 440 and 870 nm, while the turbidity β is obtained from the α value and aerosol data at 1020 nm. Since the aerosol asymmetry factor, single scattering albedo, and surface albedo are obtained at four wavelengths from AERONET in each measurement, three different spectral regions are simulated with the libRadtran model. For computations in the UV range (280-400 nm), the AERONET retrievals of aerosol asymmetry factor, aerosol single scattering albedo, and surface albedo at 440 nm are used. The AERONET retrievals at 675 nm of the same variables are used in the visible range (400-700 nm), while in the near-infrared region (700-2800 nm) we used the average properties retrieved at 870 and 1020 nm. In each interval, these properties are considered as wavelength independent. This choice to perform the radiative transfer simulations is proven as adequate in the Appendix A. Other options in the model set-up are: extraterrestrial
irradiance values are taken from Gueymard (2004); profiles of temperature, air density, ozone and other atmospheric gases are taken from the midlatitude summer/winter standard atmospheres; and the radiative equation solver is the improved version of the discrete ordinate method of Stamnes et al. (2000) (DISORT2) calculated by 16-streams (e.g., de Miguel et al., 2011). After computing the solar irradiance in the different spectral intervals, the SW irradiance is evaluated by adding up the contributions of these three spectral regions.

In order to evaluate the aerosol radiative effect, the simulations under aerosol-free conditions are also computed with the same inputs as explained above, but with a fixed $\beta$ value of 0.001.

The use of radiative transfer models fed with reliable experimental aerosol data to determine the ARE has been also employed in other studies (e.g., Barja and Antuña, 2011; Valenzuela et al., 2012; García et al., 2014).

Once the simulated radiometric values are obtained, ARE is derived for each interval (X represents UV, VIS, NIR, and SW) at the surface by:

$$\text{ARE}_X = \left( X_{\text{aer}}^\downarrow - X_{\text{aer}}^\uparrow \right) - \left( X_{\text{NOaer}}^\downarrow - X_{\text{NOaer}}^\uparrow \right) \tag{1}$$

where $X_{\text{aer}}$ and $X_{\text{NOaer}}$ are the irradiances (W m$^{-2}$) for the X range under actual and aerosol-free conditions, respectively.

Daily values are obtained by the integration of the hourly data during the whole day (24 h) considering $\text{ARE} = 0$ Wm$^{-2}$ for SZA $> 90^\circ$ (e.g., Bush and Valero, 2003; Valenzuela et al., 2012) and assuming cloud-free conditions along the day:
The aerosol forcing efficiency (AFE) is defined as the rate at which the radiative effect varies per unit of AOD (e.g., Di Biagio et al., 2009; and the references therein). The linear relationship between aerosol radiative effect and AOD is well known (see, e.g., Costa et al., 2004, 2006; Di Biagio et al., 2009). Hence, in this study, ARE is obtained as the slope of linear fits in the ARE vs AOD\textsubscript{500nm} relationships. Therefore, AFE values are expressed in W m\textsuperscript{-2} per AOD\textsubscript{500nm}-unit (Wm\textsuperscript{-2}\tau\textsuperscript{-1}).

With respect to the temporal trends calculated in this study, the Sen's method (Sen, 1968) is applied to evaluate the slope of a time series using the Mann-Kendall non parametric test to determine the significance of these rates. The Sen’s method is not greatly affected by outliers and can be computed when there are gaps in the database (Collaud Coen et al., 2013). This is a common and adequate method in temporal trend evaluation (e.g., Sánchez-Lorenzo et al., 2013). The trends calculated in this study are obtained in the corresponding physical units per year. However, to unify notation with previous studies dealing with the radiative effect trends of clouds and aerosols (e.g., Mateos et al., 2013a), the results are multiplied by 10 and expressed in physical units per decade. In this way, the trends are also easier to read.

4. Analysis of aerosol properties over the Iberian Peninsula

A direct CIMEL retrieval (AOD at 440 nm) is selected to perform the climatological analysis because the estimations of AOD\textsubscript{500nm} (used in the ARE calculations) are obtained using $\alpha$ values. Hence, we minimized the impact of other uncertainty sources in the AOD analysis. Besides, the results for AOD\textsubscript{440nm} and AOD\textsubscript{500nm} do not differ excessively. In order to identify the differences in
the aerosol climatology over the six sites analyzed in this study, the monthly distribution of the
daily values of the AOD\textsubscript{440nm} and $\alpha$ are evaluated using the database mentioned in Table 1. All the
available level 2.0 AERONET measurements are used in this section.

Figure 1 shows the climatology of the aerosol load by box whisker plots. Several conclusions can
be drawn from this figure. The highest values of the AOD occur in Barcelona, as can be expected
because it is a large city. With respect to the monthly average values (triangles in the figure), the
central stations in the Iberian Peninsula (Palencia and Évora) exhibit AOD\textsubscript{440nm} below 0.2, while the
southern sites (Cabo da Roca, Granada, and El Arenosillo) show aerosol load over 0.2 during
summer months. The AOD\textsubscript{440nm} seasonal distribution is seen, with maximum values in summer and
minimum ones in winter. However, the seasonality becomes more evident in the stations outside the
central area of the Iberian Peninsula. The large differences between median and average values for
some months evidence a large impact of high aerosol optical depth events on the monthly
climatology. In this line, the bimodality of the monthly AOD climatology (with two maximum
monthly means occurring in March and summer months) observed for the El Arenosillo site has
been already reported by previous studies (e.g., Bennouna et al., 2011), and directly attributed to
desert dust intrusions from the African continent.

To go further in the characterization, $\alpha$ allows for a better understanding of the particle size over
each site. Figure 2 shows the climatology of this variable over the six stations using also box
whisker plots. Analyzing the monthly average means, $\alpha$ values larger than one, indicative of the
predominance of fine particles, are dominant over Barcelona, Palencia, and Évora. The other three
stations (Cabo da Roca, Granada, and El Arenosillo) present monthly $\alpha$ averages over and below 1,
which means a larger variety of aerosol sizes over these stations. A seasonal dependence over
Granada site is seen, with winter months dominated by fine particles (see also Lyamani et al., 2012)
and summer months by larger ones (see also Navas-Guzman et al., 2013). Values of $\alpha$ present a
large variability during summer which is indicative of the influence of different aerosol types including biomass burning events and Saharan dust transport (e.g., Pérez-Ramírez, 2008). The monthly distribution of \( \alpha \) is symmetric with similar average and median values through the year for the six sites.

With the daily AOD and \( \alpha \) values, it is possible to classify the origin of the aerosol particles. Previous studies suggest different thresholds of AOD and \( \alpha \) (e.g., Hess et al., 1998; Pace et al., 2006; Toledano et al., 2007b). A simple classification, which can be used for the whole Iberian Peninsula, of aerosol type is carried out in this study. The threshold between fine and large particles is placed at \( \alpha = 1 \), while the situations with a high aerosol load are those with AOD\textsubscript{440nm} > 0.2. Therefore, aerosol particles can be classified in four types: maritime (AOD\textsubscript{440nm} < 0.2 and \( \alpha < 1 \)), desert dust (AOD\textsubscript{440nm} > 0.2 and \( \alpha < 1 \)), continental clean (AOD\textsubscript{440nm} < 0.2 and \( \alpha > 1 \)), and continental polluted (AOD\textsubscript{440nm} > 0.2 and \( \alpha > 1 \)). Note that the limit of AOD\textsubscript{440nm} < 0.2 is arbitrary and this value could be adjusted according to the sites, which likely produce a different distribution in the pie diagrams. Actually, even close stations can present slight differences in the \( \alpha \)-AOD classification (see, e.g., Obregón et al. 2012). However it is not the aim of this work to provide an extensive aerosol climatology, but rather to demonstrate the great variety of air masses over Iberia which transport different aerosol types. Although other types, such as biomass burning or mixed aerosols, are placed in the boundaries of these types, this simple classification can provide information about the aerosol sources for the six sites. The classification used here is line with the previous studies. For instance, Toledano et al., (2007) proposed for El Arenosillo site similar thresholds (see their Table V), although they identified continental polluted aerosols with an AOD\textsubscript{440nm} larger than 0.35 and \( \alpha > 1.4 \). Pace et al., (2006) proposed at Lampedusa island (Central Mediterranean) a desert dust identification when AOD\textsubscript{440nm} \( \geq 0.15 \) and \( \alpha \leq 0.5 \).
Figure 3 shows pie diagrams with the frequency of occurrence of the four aerosol types. The six diagrams agree pointing at continental clean as the main type of aerosols over the Iberian Peninsula. In Barcelona, there is also an important contribution of continental polluted, since Barcelona is a large coastal city with relevant pollution levels from vehicular and ship traffic (e.g., Reche et al., 2011). The influence of maritime aerosols is notable at El Arenosillo, Cabo da Roca, and Évora sites (Toledano et al., 2007a, Bennouna et al., 2011; Obregón et al., 2012). Furthermore, desert dust events are shown to be common in the Iberian Peninsula with a higher occurrence at Granada and El Arenosillo sites (the two closest points to the African continent and hence to the Saharan desert) (see also Toledano et al., 2007; Guerrero-Rascado et al., 2008; Antón et al., 2012b). For instance, the minimum values of $\alpha$ obtained for Granada station during summer months are linked to the higher likelihood of desert dust events (Valenzuela et al., 2012), being sometimes associated with high aerosol loads (Córdoba-Jabonero et al., 2011). These results corroborate the findings obtained by previous studies about desert dust events over the Iberian Peninsula (see, e.g., Lyamani et al., 2005; Cachorro et al., 2006; 2008; Toledano et al., 2007b).

The temporal trend of aerosol load can be established over the last decade in the Iberian Peninsula. The yearly values of AOD$_{440\text{nm}}$ at the six sites are shown in Figure 4. The geographical distribution of AOD through the Spanish geography is observed in the figure. Barcelona site presents yearly values over ~0.2. Granada, El Arenosillo, and Cabo da Roca exhibit yearly means in the interval between 0.15 and 0.22, while the means for Palencia and Évora sites are slightly lower in the range 0.12-0.18. Analyzing the six sites together, the year of 2010 presents one of the minimum values of AOD$_{440\text{nm}}$, while the maximum averages seem to appear at the early 2000s. The different sampling of AOD measurements in the six sites can produce discrepancies because different events are or are not captured in each database. Looking at the years with a large sampling (>200 days in, at least, four stations), 2005, 2007, and 2011, all the features mentioned above are corroborated for these particular years. The minimum of 2010 occurs when two Southern sites (El Arenosillo and Cabo da
Roca) have not enough data to evaluate the yearly mean. Hence, we cannot ensure that the apparent minimum of AOD recorded that year is linked to global-scale phenomena or to more local conditions at the other sites. During 2010 a persistent negative phases of North Atlantic Oscillation (NAO) and Quasi Biennial Oscillation (QBO) indexes was observed (e.g., Steinbrecht et al., 2011), and the connection between air mass transport at global scale and particulate matter (at the surface) is proved by Pey et al., (2013) in the Eastern Iberian Peninsula.

With respect to the temporal trend rates, the evolution of these yearly values seems to be weak. The evaluation of the trend rates (Mann-Kendall test with the 95% significance level) only produces one statistically significant trend for the Barcelona site, where a decrease of the aerosol load of 0.09 AOD$_{440 nm}$-unit per decade is observed. Although the results obtained for the other sites are not statistically significant, the sign of the temporal trends is negative for all of them. In particular, Évora and Palencia stations showed trend rates of -0.06 and -0.04 AOD$_{440 nm}$-unit per decade with significance levels of 94% and 90%, respectively. Hence, a slight reduction of the aerosol load over the Iberian Peninsula is observed since 2000. This result obtained in the Southeastern Europe is in line with the long-term analysis of AOD series performed in Northern Germany and Switzerland by Ruckstuhl et al. (2008). These authors highlight a strong decrease of aerosol load starting in 1985, while the values are stabilized since about 2000.

The reasons behind the decrease in the aerosol load since the early 2000s are a mixed of anthropogenic and natural sources. As was reported by Aas et al. (2013), the particulate matter (PM) emissions in the Iberian Peninsula have decreased around 25% between 2000 and 2011. Furthermore, observational PM data in different Spanish sites have also shown a decrease trend in the 2000s (e.g., Barmpadimos et al., 2012; Cusack et al., 2012; Pey et al., 2013; Bennouna et al., 2014). This fact can be understood by the effect of the current economic crisis and the implementation of new environmental laws to control the pollution (e.g., Querol et al., 2014). In
addition, recent studies have shown that natural aerosols have also decreased in the last decade. For instance, Gkikas et al. (2013) reported, using satellite AOD estimations, that strong and extreme desert dust episodes in the Mediterranean decreased in the period 2000-2007 over land surfaces. This trend is understood due to the low spring and summer frequencies in 2005 and 2007 and the high frequencies in 2000 and 2003. As it was shown by Pey et al. (2013), one possible reason behind this trend is the atypical trajectories followed by the air masses emerging from North Africa in summer since 2006. Hence, both columnar and surface aerosols have pointed out a decrease in the aerosol load concerning both anthropogenic and natural aerosols.

5. Inter-annual and intra-annual evolution of ARE

From the daily data, the yearly ARE for each station and spectral range are evaluated to analyze their inter-annual changes (see Figure 5). In spite of the high variability of the yearly values with large standard deviations (see the vertical bars for Palencia station in the figure), the radiative effects of atmospheric aerosols have slightly declined over the last years. The patterns of ARE in the UV, VIS, NIR, and SW ranges are similar, since the inter-annual changes are simultaneously observed in the four spectral intervals. The significance levels of the temporal trends (Mann-Kendall nonparametric test at the 95% confidence interval) are evaluated. Évora and Palencia sites exhibit statistically significant trends in the periods 2005-2012 and 2003-2012, respectively. The trends for the aerosol effects for Palencia (Évora) are: +4.9 (+3.2) Wm^{-2} per decade in \( \text{ARE}_{\text{SW}} \), +3.3 (+2.1) Wm^{-2} per decade in \( \text{ARE}_{\text{VIS}} \), +0.1 (+0.08) Wm^{-2} per decade in \( \text{ARE}_{\text{NIR}} \), and +0.06 (+0.03) Wm^{-2} per decade in \( \text{ARE}_{\text{UV}} \). The other four stations present positive trends in all the spectral ranges, but they are not statistically significant at the 95% confidence interval. The decrease in the aerosol effects is in line with the previous fall in the AOD values in the 2000s mentioned in Section 4. Furthermore, this slight reduction in the radiative effects of the atmospheric aerosol over the Iberian
Peninsula could partially contribute to the increase in the levels of SW radiation at the surface (the brightening phenomenon) in this region reported by Sanchez-Lorenzo et al. (2013) and Mateos et al. (2013a).

To establish the general behavior of the AR E over the whole Iberian Peninsula, the yearly values using the six ground-based stations are evaluated. Only those years with, at least, simultaneous measurements at three sites are considered in these averages, and consequently, the time period is limited to 2004-2012. Figure 6 shows the evolution of the ARE and AOD at 500 nm for the entire peninsula. The previously discussed reduction of aerosol load in the six individual datasets is again corroborated with the mean data series. This decline produces a consequent decrease in the aerosol radiative effect at the four spectral ranges. The temporal trends of these yearly values are evaluated, and all the trends resulted statistically significant at the 95% significance level are shown in Figure 6. Overall, AR_{SW} over the Iberian Peninsula increased 3.6 W m^{-2} per decade while the aerosol reduced 0.04 AOD_{500nm-unit} per decade. The yearly aerosol radiative effects over the entire peninsula are in the ranges: \(-1.1 < \text{AR}_{\text{EUV}} < -0.7 \) W m\(^{-2}\), \(-5.7 < \text{AR}_{\text{VIS}} < -3.5 \) W m\(^{-2}\), \(-2.6 < \text{AR}_{\text{NIR}} < -1.6 \) W m\(^{-2}\), and \(-8.8 < \text{AR}_{\text{EWS}} < -5.7 \) W m\(^{-2}\). The larger contribution of the visible spectral region with respect to the whole solar spectrum was also noticed by Bush and Valero (2003). The relationship between ARE and AOD_{500nm} is analyzed more in detail in Section 6, when the aerosol forcing efficiency is evaluated for each ground-based station.

In addition to the inter-annual changes, the intra-annual behavior is also analyzed. For this purpose, the annual cycle (12 monthly means) is evaluated for the six stations (see Figure 7). A seasonal pattern is seen in \text{AR}_{\text{EUV}} and \text{AR}_{\text{VIS}}, and therefore, \text{AR}_{\text{SW}}. However, \text{AR}_{\text{NIR}} does not follow a seasonal pattern, particularly at the Évora and Palencia stations given that \text{AR}_{\text{NIR}} remains nearly constant. Small differences among the six stations are observed in the annual cycle during the cold seasons. The aerosol radiative effects are stronger during summer months. This can be related to the
higher likelihood of desert dust or biomass burning events over the Iberian Peninsula in these months (e.g., Cachorro et al., 2008; Valenzuela et al., 2012), as was mentioned above. This is corroborated by the increase of the differences among the stations during the warm season, likely due to the variability in the impact of the desert dust episodes which strongly depend on the geographical location of each site. The higher occurrence of large aerosol loads during the warm seasons (see Figure 1), can explain the more negative ARE during summer and spring in Figure 7. For instance, the Barcelona station, with the largest values of AOD_{440nm}, is the bottom curve of each panel in Figure 7. Furthermore, the influence of mineral dust aerosol (with high aerosol optical depth) during these months also causes strong radiative effects, as was also reported by previous studies (e.g., Cachorro et al., 2008; Guerrero-Rascado et al., 2009; Antón et al., 2012a, 2012b; Román et al., 2013; García et al., 2014). In addition, the bimodality of the monthly AOD climatology mentioned in Section 4 has its impact on the radiative effects. The annual AOD cycle (see Figure 1, El Arenosillo site) causes the inverse monthly distribution of ARE with a first minimum in March. This effect is more clearly seen in ARE_{NIR} and ARE_{SW}.

6. Aerosol radiative forcing efficiency in different spectral ranges

The daily AFE values are calculated (following the methodology described in Section 3) in all the spectral ranges. AFE is a function of the aerosol optical properties, where both the aerosol particle size distribution and absorptive properties play a key role (e.g., Antón et al., 2011). As we assumed a fixed value of SSA = 0.90 in the simulations with AOD_{440nm} < 0.15 (see Table 2), the AFE is calculated only for those cases showing AOD_{440nm} larger than 0.15.

To identify the influence of SSA and \( \alpha \) on AFE, this variable is calculated for several intervals of each aerosol property. Four categories of single scattering albedo at 675 nm are established in the calculation of the AFE: \( 1.0 \geq SSA_1 > 0.95, \ 0.95 \geq SSA_2 > 0.90, \ 0.90 \geq SSA_3 > 0.85, \) and \( 0.85 \geq SSA_4 > 0.80 \).
Furthermore, aerosol size is classified in three intervals: $0 \leq \alpha_1 \leq 1$, $1 < \alpha_2 \leq 1.5$, and $1.5 < \alpha_3 \leq 2$. Note that two intervals in the range of $\alpha$ larger than 1 have been considered. One for median particles and another one for fine particles, because of the relevant importance of median size particle (continental or mixed aerosol aerosols types) over the Iberian Peninsula (see Figure 3).

Although the general classification between fine and coarse particles requires a more refined classification (Schuster et al., 2006; Prats et al., 2011), the more general intervals selected in this study are adequate to perform a study of the aerosol sizes at the six stations together.

Figure 8 shows the AFE obtained for the UV (AFE$_{UV}$), VIS (AFE$_{VIS}$), NIR (AFE$_{NIR}$), and SW (AFE$_{SW}$) ranges for all these intervals. The threshold to evaluate the average in each sub-interval is fixed at 10 data points. From these figures it is seen that, the stronger the absorption by aerosols, the stronger their forcing efficiency. That is a decrease in the absolute values of the AFE for increasing SSA and for all particle size. In general, the groups of non-absorbing particles exhibit a good agreement among the six stations (see, for instance, AFE values in all the spectral ranges in the interval $1 < \alpha \leq 1.5$). Larger differences are obtained in the case of more absorbing aerosol particles. These can be understood because of the different types of aerosols presented over each site (see Section 4) and the different data numbers. The average AFE values over the whole Iberian Peninsula (considering the six stations together) are presented in Table 3 as a function of $\alpha$ and SSA, separately. The role played by the aerosol size on AFE values is different in the three sub-intervals of the shortwave radiation. AFE$_{UV}$ and AFE$_{VIS}$ are larger (in absolute value) for fine particles, while the opposite occurs in the case of AFE$_{NIR}$. As a result of these mixed effects, AFE$_{SW}$ shows also a decrease in its values with increasing $\alpha$, but this effect is weaker than for the visible and ultraviolet parts. SSA exhibits a more dominant role. As was observed before, the most negative values are achieved for the most absorbing aerosols considered in this study (group 1 of SSA, see Table 3).
The average values of forcing efficiency obtained in this study (see Table 3) are in line with those found by other authors. Table 4 summarizes the results obtained by previous studies. It is difficult to assess some features in the comparison with previous reported AFE values, because of the different aerosol types, time periods and methods that are analyzed. Our study presents the evaluation of ARE with six long-term databases of aerosol properties. In spite of that, the values shown in Table 3 agree with those of Table 4, but the larger discrepancies are observed with the studies focusing on specific events. Our results match better with the results reported by, e.g., Zhou et al. (2005), Meloni et al. (2005), and Di Biagio et al. (2010). As was noticed by, e.g., Costa et al. (2004, 2006) and Di Biagio et al. (2010), AFE at the surface is larger (in absolute term) for aerosols characterized by smaller and absorbing particles. This result is corroborated by the findings shown in this study. Furthermore, as was pointed out by Di Biagio et al. (2010), the aerosol absorption is the dominant factor on AFE evaluated at the surface.

To evaluate the contribution of each spectral range with respect to the shortwave, the dependence of each AFE ratio (VIS to SW and NIR to SW) on SSA and α is shown in Figure 9. AFEVIS/AFE_{SW} and AFE_{NIR}/AFE_{SW} ratios are shown in the figure since their contributions are the dominant. AFE_{UV}/AFE_{SW} ratio can be obtained as 100% minus the sum of the percentage of the two other ranges. As expected, non substantial differences are observed in the behavior of the six stations considered in this study. The NIR contribution becomes more decisive for large particles (α < 1). It is expected that larger particles interact more with the longer wavelengths, while the smaller particles present more interaction with the shorter wavelengths. The presence of large particles with low SSA (high absorption) leads to a reduction of the AFE_{NIR}/AFE_{SW} ratio as well as an increase of the AFE_{VIS}/AFE_{SW} ratio. However, for non-absorbing (high SSA) large particles, the AFE_{NIR}/AFE_{SW} ratio increases, and the contributions of the visible and infrared parts become more similar (both around ~40-50%). The difference between AFE_{VIS}/AFE_{SW} and AFE_{NIR}/AFE_{SW} increases for intermediate - fine particles. For these particles, the AFE_{VIS}/AFE_{SW} ratio does not
show a dependence on SSA. The smallest contribution of the NIR interval is around ~25% under strong absorbing aerosols and fine particles, while $\text{AFE}_{\text{VIS}}/\text{AFE}_{\text{SW}}$ is still over 60%. For this case, the contribution of the ultraviolet range achieves a maximum of ~15%, being almost comparable with the near infrared contribution. In summary, aerosol size determines the relevance of VIS-NIR ranges, while SSA plays a key role, particularly, for large particles.

7. Conclusions

Six long-term datasets of aerosol properties over the Iberian Peninsula were analyzed and used as input in a radiative transfer model to simulate ultraviolet, visible, near-infrared, and shortwave radiation. The aerosol radiative effect (ARE) and aerosol forcing efficiency (AFE) were calculated. The main conclusions are as follows:

1) The annual cycles of AOD and $\alpha$ values of atmospheric aerosols over the six analyzed stations present high variability among them, emphasizing the inhomogeneity of the Iberian Peninsula, mainly due to the different aerosol types over each station. The Barcelona site presents the largest values of AOD, although Southern stations (Granada and El Arenosillo sites) frequently exhibit daily values over 0.2 during summer months. The classification $\alpha$-AOD has shown that continental (mainly, clean) is the principal type of aerosol over the Iberian Peninsula. However, maritime aerosols are also common in the Cabo da Roca, El Arenosillo and Évora sites. Desert dust events are registered at the six sites, with the highest frequency at Granada and El Arenosillo, but the most relevant feature is the South-North gradient of desert dust load which modulates the aerosol climatology over the Iberian Peninsula.

2) The aerosol load over the Iberian Peninsula has shown a decrease trend between 2004 and 2012 (-0.04 per unit of AOD$_{500\text{nm}}$ per decade, being statistically significant at the 95% of significance
level). Yearly values of the AOD at 440 nm have also shown a statistically significant trend of -0.09 AOD_{440nm}\text{-unit per decade at Barcelona site. The temporal trends for the rest of the stations are not statistically significant at the 95% significance level, but all of them are negative. Hence, a reduction of the aerosol column load over the Iberian Peninsula is observed in the last decade.

3) In the whole Iberian Peninsula, yearly ARE_{UV} ranges between -1.1 and -0.7 Wm^{-2}, ARE_{VIS} ranges between -5.7 and -3.6 Wm^{-2}, and ARE_{NIR} has values between -2.6 and -1.6 Wm^{-2}. As a result, ARE_{SW} is in the range between -8.8 and -5.7 Wm^{-2}. The temporal trends of ARE_{UV}, ARE_{VIS}, ARE_{NIR}, and ARE_{SW} exhibit positive statistically significant trends between 2004 and 2012. For instance, the trend rate for the ARE_{SW} is +3.6 Wm^{-2} per decade (statistically significant at the 95% of significance level).

4) The intra-annual ARE cycle exhibits larger values during the spring and summer months when the likelihood of high aerosol loading over the Iberian Peninsula increases. In general, the annual AOD cycle is driven by the occurrence of Saharan dust events.

5) The AFE values at the six stations used in this study are in good agreement. Conditions of high $\alpha$ (small particles predominate) and low SSA (high absorption) lead to the largest negative AFE values. Overall, as an average for the Iberian Peninsula: $AFE_{UV} = -6 \text{ Wm}^{-2} \tau^{-1}$, $AFE_{VIS} = -34 \text{ Wm}^{-2} \tau^{-1}$, $AFE_{NIR} = -19 \text{ Wm}^{-2} \tau^{-1}$, and $AFE_{SW} = -59 \text{ Wm}^{-2} \tau^{-1}$.

6) The contribution of the ultraviolet, visible, and infrared to total shortwave aerosol forcing efficiency is governed by the aerosol type. In general, the visible part of the spectrum is the most dominant part. Non-absorbing large particles cause a more equal contribution of VIS and NIR intervals, while the UV range shows a higher contribution for absorbing fine particles.
Appendix A

The two choices in the performance of radiative transfer simulations from the libRadtran code concerning aerosol properties are justified in this section.

First at all, as it is mentioned in the text, most of the data present $\text{AOD}_{440\text{nm}} < 0.15$ (~70% for Palencia, Granada, and Évora sites). For these low values, $\text{SSA} = 0.9$ and $g = 0.75$ are selected by the representativeness of the local aerosols in the six sites of study. To analyze possible uncertainties emerging from this choice, Table A1 shows the relative difference between two extreme cases of SSA ($\text{SSA}_1 = 0.8$ and $\text{SSA}_2 = 1.0$) for different values of the asymmetry factor and two different SZA s (30° and 60°). The two optical properties are also fixed as non-wavelength-dependent in this analysis. The $\text{AOD}_{440\text{nm}}$ used is 0.15, the worst scenario possible for these cases because the higher the AOD the stronger the impact of aerosol properties. The simulations are performed for the four spectral ranges, and the net flux at the surface is evaluated for all of them. The change of SSA between the two extreme cases produce a relative difference in the SW range less than 5%, the relative difference is higher for the UV range but also <10%. Furthermore, as the SSA influences the diffuse radiation, the worst results are obtained at large SZA s. The impact of $g$ on the net fluxes is very weak. In conclusion, the choice of SSA = 0.9 and $g = 0.75$ in a clean scenario ($\text{AOD}_{440\text{nm}} < 0.15$) is proven as adequate because of two reasons: a) representativeness of the local aerosols which can be mixture of different types, and b) the low uncertainty produced in the simulations by SSA and $g$ under these conditions.

The choice of fixed SSA and $g$ values within each of the spectral ranges (UV, VIS, and NIR) represented by the CIMEL spectral measurements is also justified here. The aerosol models by Shettle (1989) included in the libRadtran code (see Mayer and Kylling, 2005) are used to evaluate the uncertainty of using this approximation. The continental clean aerosols (most common type in the Iberian Peninsula, see Figure 3), and continental polluted aerosols (also very common, which...
present an extreme case of absorption) are tested in this analysis. The simulations are performed for
the expected spectral behavior of SSA and g following Shettle (1989) and the case of fixed
properties in the UV (SSA and g values at 440 nm), VIS (SSA and g values at 675 nm), and NIR
intervals (SSA and g average of values at 870 and 1020 nm). Figure A1 presents the evolution of
the relative error (considering as reference the net flux with the expected spectral dependence of
aerosol properties) for several AOD values between aerosol-free and \( \text{AOD}_{550\text{nm}} = 0.6 \). In the case of
continental clean aerosols (Figure A1.a), the error of using our assumption is lower than 5% for all
SZAs and spectral ranges. Therefore, as the large majority of aerosol particles are of this type, the
methodology used and proposed in this study only introduces a relative error below 5% in the
majority of the simulations. With respect to the continental polluted aerosols (Figure A1.b), the
error increases achieving a maximum around 20% for the UV range and very turbid conditions. For
large AOD conditions in the Iberian Peninsula (e.g., \( \text{AOD}_{550\text{nm}} = 0.4 \)) but with low frequency of
occurrence in contrast to \( \text{AOD}_{440\text{nm}} < 0.15 \), the error of the SW range is below 5%. However, the
UV range is more sensitive to our method and the error is around 15% at SZA = 60°. As it was
mentioned above, the errors are larger for large SZAs because of the possible interaction between
absorption and scattering processes resulting the diffuse radiation. The visible range is more
sensitive to the spectral variations than the NIR interval, which exhibits a maximum error around
11% at SZA = 60° and \( \text{AOD}_{550\text{nm}} = 0.6 \). The daily net radiative fluxes are also evaluated for the two
aerosol types in order to quantify the uncertainty in the final simulated data used in this study. For
Palencia site (and the corresponding SZA evolution), a daily value for the June 20th is simulated
assuming TOC = 300 DU and PWC = 1 cm. The results for the continental polluted case with
\( \text{AOD}_{440\text{nm}} = 0.4 \) exhibit differences between the spectral and fixed-band aerosol properties of: 7.5%,
5.3%, 4.0%, and 4.8% for the UV, VIS, NIR, and SW intervals. The relative errors for the same
intervals with continental clean (and same AOD value) are: 1.9%, 1.2%, 1.4%, and 1.4%,
respectively. Therefore, the uncertainty due to fixed optical properties in each spectral range is
dependent on the aerosol type but the error caused can be considered as acceptable. Since actual
aerosols often present mixtures of different types, the uncertainty of using the theoretical spectral
evolution for one type (given by an aerosol model) can also produce uncertainties which should be
taken into account. Although other aerosol types are not tested in this analysis, a similar behavior
can be expected. For instance, for the case of desert dust aerosols, Román et al. (2013) found a
slight influence of spectral aerosol absorption properties on UV irradiance analyzing a strong
Saharan intrusion over Granada site.

Therefore, the two assumptions performed in this study in the simulations are adequate for the
evaluation of net fluxes and aerosol radiative effects. The uncertainties that can be introduced in the
daily values are acceptable being around or smaller than 5% for the net SW radiation. This
uncertainty is usually achieved in clear-sky modeling (e.g., Mateos et al., 2010; Bilbao et al., 2011).

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Table 1. Coordinates and time interval of the six AERONET sites used in this study.

<table>
<thead>
<tr>
<th>Station</th>
<th>Latitude (°N)</th>
<th>Longitude (°E)</th>
<th>Altitude a.s.l. (m)</th>
<th>Time interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>Palencia</td>
<td>41.99</td>
<td>-4.52</td>
<td>750</td>
<td>2003-2012</td>
</tr>
<tr>
<td>Barcelona</td>
<td>41.39</td>
<td>2.12</td>
<td>125</td>
<td>2004-2012</td>
</tr>
<tr>
<td>Cabo da Roca</td>
<td>38.78</td>
<td>-9.50</td>
<td>140</td>
<td>2003-2011</td>
</tr>
<tr>
<td>Évora</td>
<td>38.57</td>
<td>-7.91</td>
<td>293</td>
<td>2005-2012</td>
</tr>
<tr>
<td>Granada</td>
<td>37.16</td>
<td>-3.61</td>
<td>680</td>
<td>2004-2012</td>
</tr>
<tr>
<td>El Arenosillo</td>
<td>37.11</td>
<td>-6.73</td>
<td>0</td>
<td>2000-2009</td>
</tr>
</tbody>
</table>

Table 2. Summary of AERONET data used for ARE calculations: aerosol optical depth (AOD), single scattering albedo (SSA), asymmetry factor (g), precipitable water vapor column (PWC). Estimated absolute uncertainty of AOD and SSA is given according to Dubovik et al. (2002), and PWC error from Holben et al. (1998).

<table>
<thead>
<tr>
<th>AERONET database</th>
<th>Estimated uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>AOD</td>
<td>Level 2.0</td>
</tr>
<tr>
<td>SSA, g (AOD_{440}&gt;0.4)</td>
<td>Level 2.0</td>
</tr>
<tr>
<td>SSA, g (0.15&lt;AOD_{440}&lt;0.4)</td>
<td>Level 1.5-filtered*</td>
</tr>
<tr>
<td>SSA, g (AOD_{440}&lt;0.15)</td>
<td>Fixed value</td>
</tr>
<tr>
<td>PWC</td>
<td>Level 2.0</td>
</tr>
</tbody>
</table>

*Filters applied are the same as in level 2.0 except for AOD_{440} (see text).

Table 3. AFE values and their standard error for the UV, VIS, NIR, and SW ranges for, separately, four SSA and three α intervals over the Iberian Peninsula. Units are Wm^{-2}τ^{-1}. SSA groups: 0.85 ≥ SSA_{1} > 0.80 (group 1), 0.90 ≥ SSA_{2} > 0.85 (group 2), 0.95 ≥ SSA_{3} > 0.90 (group 3), and 1.0 ≥ SSA_{4} > 0.95 (group 4); and α groups: 0 ≤ α_{1} ≤ 1 (group 1), 1.0 ≤ α_{2} ≤ 1.5 (group 2), and 1.5 < α_{3} ≤ 2 (group 3). The average values without any classification are also presented.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Group</th>
<th>AFE_{UV}</th>
<th>AFE_{VIS}</th>
<th>AFE_{NIR}</th>
<th>AFE_{SW}</th>
</tr>
</thead>
<tbody>
<tr>
<td>α</td>
<td>1</td>
<td>-5.41 ± 0.06</td>
<td>-30.1 ± 0.3</td>
<td>-20.9 ± 0.2</td>
<td>-56.5 ± 0.5</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>-6.60 ± 0.09</td>
<td>-38.3 ± 0.4</td>
<td>-19.1 ± 0.2</td>
<td>-64.0 ± 0.6</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>-7.06 ± 0.10</td>
<td>-39.4 ± 0.4</td>
<td>-16.9 ± 0.2</td>
<td>-63.3 ± 0.7</td>
</tr>
<tr>
<td>SSA</td>
<td>1</td>
<td>-9.7 ± 0.2</td>
<td>-52.8 ± 0.8</td>
<td>-24.9 ± 0.5</td>
<td>-87.4 ± 1.4</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>-8.19 ± 0.10</td>
<td>-44.6 ± 0.4</td>
<td>-21.2 ± 0.2</td>
<td>-74.0 ± 0.6</td>
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<td>3</td>
<td>-6.37 ± 0.05</td>
<td>-35.9 ± 0.2</td>
<td>-19.5 ± 0.2</td>
<td>-61.8 ± 0.3</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>-4.59 ± 0.05</td>
<td>-26.6 ± 0.2</td>
<td>-18.1 ± 0.2</td>
<td>-49.3 ± 0.3</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td>-5.98 ± 0.05</td>
<td>-33.7 ± 0.2</td>
<td>-19.34 ± 0.11</td>
<td>-59.1 ± 0.3</td>
</tr>
</tbody>
</table>
Table 4. Daily Forcing Efficiencies at the surface by previous studies. Legend: desert dust (DD), continental-biomass burning (C-BB), and forest fires (FF).

<table>
<thead>
<tr>
<th>Reference</th>
<th>Aerosol Type</th>
<th>AFE&lt;sub&gt;X&lt;/sub&gt;</th>
<th>Value (Wm&lt;sup&gt;-2&lt;/sup&gt;τ&lt;sup&gt;-1&lt;/sup&gt;)</th>
<th>Time period</th>
<th>Region</th>
<th>More info.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diaz et al. (2007)</td>
<td>Mixed</td>
<td>AFE&lt;sub&gt;UV&lt;/sub&gt;</td>
<td>-3</td>
<td>July 2002</td>
<td>Spain</td>
<td>290-363 nm</td>
</tr>
<tr>
<td>Meloni et al. (2005)</td>
<td>DD Mixed</td>
<td>AFE&lt;sub&gt;VIS&lt;/sub&gt;</td>
<td>-28.4</td>
<td>July 2002</td>
<td>Central Mediterranean</td>
<td></td>
</tr>
<tr>
<td>Lyamani et al. (2006)</td>
<td>Mixed</td>
<td>AFE&lt;sub&gt;VIS&lt;/sub&gt;</td>
<td>-75.8</td>
<td>August 2003</td>
<td>Spain</td>
<td>2003 heat wave</td>
</tr>
<tr>
<td>Di Biagio et al. (2010)</td>
<td>C-BB Mixed</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-68.9</td>
<td>2004-2007</td>
<td>Central Mediterranean</td>
<td>At the equinox</td>
</tr>
<tr>
<td>Esteve et al. (2014)</td>
<td>Mixed</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-139.0</td>
<td>2003-2011</td>
<td>Spain</td>
<td>200 cloud-free days</td>
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<tr>
<td>Santos et al. (2008)</td>
<td>FF</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-113.0</td>
<td>2004-2005</td>
<td>Portugal</td>
<td>Absorbing aerosols</td>
</tr>
<tr>
<td>di Sarra et al. (2011)</td>
<td>DD</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-55</td>
<td>25-26/03/2010</td>
<td>Central Mediterranean</td>
<td>Strong event</td>
</tr>
<tr>
<td>Garcia et al. (2014)</td>
<td>DD</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-59</td>
<td>2009-2012</td>
<td>Canary Islands</td>
<td>386 cloud-free days</td>
</tr>
<tr>
<td>Costa et al. (2006)</td>
<td>DD</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-116.9</td>
<td>7/04/2000</td>
<td>Korea</td>
<td>SSA = 0.76</td>
</tr>
<tr>
<td>Zhou et al. (2005)</td>
<td>DD</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-80/48</td>
<td>Monthly aerosol climatology</td>
<td>North Africa and Arabian Peninsula</td>
<td>Depending on surface albedo</td>
</tr>
<tr>
<td>Saha et al. (2008)</td>
<td>C-BB</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-97.6</td>
<td>2005-2006</td>
<td>French Mediterranean</td>
<td>0.7 &lt; SSA &lt; 0.8</td>
</tr>
<tr>
<td>Valenzuela et al. (2012)</td>
<td>DD</td>
<td>AFE&lt;sub&gt;SW&lt;/sub&gt;</td>
<td>-70</td>
<td>2005-2010</td>
<td>Spain</td>
<td></td>
</tr>
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</table>

Table A1. Relative difference (RD) in the UV, VIS, NIR, and SW net fluxes between the case of SSA<sub>1</sub>=0.8 and SSA<sub>2</sub>=1.0 for different g and SZA values.

<table>
<thead>
<tr>
<th>SZA</th>
<th>g</th>
<th>RD&lt;sub&gt;UV&lt;/sub&gt; (%)</th>
<th>RD&lt;sub&gt;VIS&lt;/sub&gt; (%)</th>
<th>RD&lt;sub&gt;NIR&lt;/sub&gt; (%)</th>
<th>RD&lt;sub&gt;SW&lt;/sub&gt; (%)</th>
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<td>30º</td>
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<td>-1.8</td>
<td>-3.0</td>
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<tr>
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<td>-6.5</td>
<td>-3.7</td>
<td>-1.8</td>
<td>-3.0</td>
</tr>
<tr>
<td></td>
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<td>-3.8</td>
<td>-1.9</td>
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<tr>
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<tr>
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<td>-9.6</td>
<td>-6.0</td>
<td>-3.0</td>
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Figure Captions

Figure 1. Annual cycle of daily values of AOD at 440 nm by box whisker plots. Triangles and horizontal solid lines indicate the monthly average and median values, respectively.

Figure 2. Annual cycle of daily values of $\alpha$ ('alpha' in the figure) by box whisker plots. Triangles and horizontal solid lines indicate the monthly average and median values, respectively.

Figure 3. Relative frequency of aerosol type occurrence: maritime (MA), desert dust (DD), continental clean (CC), and continental polluted (CP).

Figure 4. Yearly values of AOD$_{440\text{nm}}$ at the six sites: Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El Arenosillo (green circles). The text points out the statistically significant trend obtained.

Figure 5. Evolution of yearly ARE$_{\text{UV}}$ (a), ARE$_{\text{VIS}}$ (b), ARE$_{\text{NIR}}$ (c), and ARE$_{\text{SW}}$ (d) at the six sites: Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El Arenosillo (green circles). Vertical bars indicate the standard deviation of each yearly value at Palencia station.

Figure 6. Evolution of annual ARE at the four spectral ranges (ARE$_{\text{UV}}$ purple diamonds, ARE$_{\text{VIS}}$ red squares, ARE$_{\text{NIR}}$ green triangles, and ARE$_{\text{SW}}$ black circles) and AOD at 500 nm (blue stars) averaging the data from the six Iberian ground-based sites (only years with at least three sites considered). Dashed lines point out the linear trends (see text).

Figure 7. Annual cycle of ARE$_{\text{UV}}$ (a), ARE$_{\text{VIS}}$ (b), ARE$_{\text{NIR}}$ (c), and ARE$_{\text{SW}}$ (d) at the six sites: Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El Arenosillo (green circles). Vertical bars point out the standard deviation of each monthly value at Évora station.
Figure 8. AFE_{UV}, AFE_{VIS}, AFE_{NIR}, and AFE_{SW} against four groups of aerosol single scattering albedo and three intervals of $\alpha$ at the six sites: Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El Arenosillo (green circles).

Figure 9. Dependence of AFE_{VIS}/AFE_{SW} (a, c, e) and AFE_{NIR}/AFE_{SW} (b, d, f) ratios on SSA for large (a, b), medium (c, d) and small (e, f) particles at the six sites: Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El Arenosillo (green circles).
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Figure A1. Evolution of the error committed when the optical properties are fixed in the different spectral range for two SZAs, and continental clean (CC, a) and continental polluted (CP, b) aerosols.