

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

D. Mateos<sup>1,2</sup>, M. Antón<sup>1</sup>, C. Toledano<sup>2</sup>, V. E. Cachorro<sup>2</sup>, L. Alados-Arboledas<sup>3,4</sup>, M. Sorribas<sup>3,4,5</sup>, M.J. Costa<sup>6</sup>, J.M. Baldasano<sup>7</sup>.

1. Departamento de Física, Universidad de Extremadura, Badajoz, Spain

2. Grupo de Óptica Atmosférica, Universidad de Valladolid, Valladolid, Spain

3. Departamento de Física Aplicada, Universidad de Granada, Granada, Spain

4. Andalusian Institute for Earth System Research, Universidad de Granada, Granada, Spain

5. Estación de Sondeos Atmosféricos El Arenosillo, INTA, Huelva, Spain

6. Évora Geophysics Centre and Dep. Physics, University of Évora, Évora, Portugal

7. Universidad Politécnica de Cataluña, Barcelona, Spain

Corresponding author: D. Mateos, Grupo de Óptica Atmosférica, Universidad de Valladolid, Paseo Belén 7, 47011, Valladolid, Spain. E-mail: mateos@goa.uva.es

## 20 Abstract

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

43

## 44    **1. Introduction**

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

55    The aerosol radiative effect (ARE) is defined as the change in net radiation due to changes in  
56    atmospheric aerosol properties and content. This is a key quantity in the determination of climate  
57    change (e.g., Hansen et al., 1998). Most studies dealing with ARE have focused on discrete  
58    wavelengths, whole shortwave (SW) solar radiation spectrum (e.g., Rajeev and Ramanathan, 2001;  
59    García et al., 2008; di Sarra et al., 2008; Cachorro et al., 2008; Foyo et al., 2014), longwave (LW)  
60    radiation (e.g., Panicker et al., 2008; di Sarra et al., 2011; Antón et al., 2014), ultraviolet (UV)  
61    interval (e.g., Hatzianastassiou et al., 2004; Kazadzis et al., 2009; Nikitidou et al., 2013), and visible  
62    (VIS) range (e.g., Jayaraman et al., 1998; Horvath et al., 2002; Bush and Valero, 2003; Meloni et  
63    al., 2003). With regards to surface SW radiative effect ( $ARE_{SW}$ ), Di Biagio et al. (2010) obtained  
64    the maximum radiative daily effects for different aerosol types in the Central Mediterranean in the  
65    period 2004-2007:  $-61 \text{ Wm}^{-2}$  (desert dust aerosols),  $-26 \text{ Wm}^{-2}$  (urban/industrial - biomass burning  
66    aerosols) and  $-43 \text{ Wm}^{-2}$  (mixed aerosols). All these negative figures point out a cooling of the  
67    Earth's surface. Aerosol radiative effects in the LW ( $ARE_{LW}$ ) are expected to be smaller than in the

68 SW and with positive sign (see, e.g., di Sarra et al., 2011; Antón et al., 2014). Hence, this heating  
69 effect at the surface can partly offset the cooling induced in the SW range. With respect to the ARE  
70 for the UV range ( $ARE_{UV}$ ), Nikitidou et al. (2013) analyzed the ARE in two different spectral  
71 regions in the UV range, 300-315 and 315-360 nm. They found a stronger attenuation in the UV-B  
72 than in the UV-A, however being both solar zenith angle (SZA) dependent because of different  
73 photon paths.

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

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

92

## 93    **2. Columnar aerosol optical data**

94    The aerosol data are obtained from the Aerosol Robotic Network (AERONET) (Holben et al.,  
95    1998). Six AERONET sites operating in the Iberian Peninsula are selected in this study: Palencia,  
96    Barcelona, Évora, Cabo da Roca, Granada and El Arenosillo (see Table 1), all of them with a  
97    minimum of 8 years of data sets of continuous observations. These sites present the largest records  
98    of aerosol properties in the Iberian Peninsula in the AERONET network.

99    The standard instrument used in AERONET is the Cimel CE-318 radiometer. It performs direct sun  
100    measurements at several wavelengths in the spectral range 340-1020 nm. Furthermore, the  
101    instrument also measures sky radiance in the solar almucantar and principal plane configurations at  
102    440, 670, 870 and 1020 nm wavelengths. A detailed description of this instrument was provided by  
103    Holben et al. (1998). The direct sun observations are used to derive the spectral aerosol optical  
104    depth (AOD) and the corresponding Ångström exponent. The sky radiances together with the AOD  
105    are employed to retrieve a set of aerosol optical and microphysical properties via inversion methods  
106    (Dubovik and King, 2000; Dubovik et al., 2006). These include particle size distribution, complex  
107    refractive index, single scattering albedo (SSA), phase function, asymmetry parameter, fraction of  
108    non-spherical particles, etc. (see  
109    [http://aeronet.gsfc.nasa.gov/new\\_web/Documents/Inversion\\_products\\_V2.pdf](http://aeronet.gsfc.nasa.gov/new_web/Documents/Inversion_products_V2.pdf)). Data are provided  
110    in three database levels: 1.0 (raw data), 1.5 (cloud-screened) and 2.0 (cloud-screened and quality  
111    assured).

112    The calibration of these instruments is performed following AERONET protocols by AERONET-  
113    NASA, PHOTONS and RIMA networks every 12 months of operation approximately. The  
114    estimated uncertainty is 0.01-0.02 for AOD (larger at shorter wavelengths) and ~5% for the sky

115 radiances (Holben et al., 1998). The SSA has an absolute uncertainty about 0.03-0.07 depending on  
116 the aerosol load and type (Dubovik et al., 2000).

117 Level 2.0 aerosol optical depth data have been used in this work. However, it is well-known that  
118 when level 2.0 inversion data are used, the number of available observations of single scattering  
119 albedo (SSA) and asymmetry factor ( $g$ ) is quite limited because these variables are only considered  
120 reliable when  $AOD_{440nm} > 0.4$ <sup>1</sup>. Such AOD is mainly reached in the study region during Saharan  
121 dust or biomass burning events, therefore we would not have information on SSA and  $g$  for other  
122 conditions. To solve this issue, we have reduced the threshold of the level 2.0 inversion products.  
123 For this, we started with the level 1.5 data (for those quality-assured almucantar data that reached  
124 level 2.0) and applied the same criteria used by AERONET to elaborate the level 2.0 regarding the  
125 number of symmetrical angles, retrieval error and solar zenith angle (see  
126 [http://aeronet.gsfc.nasa.gov/new\\_web/Documents/AERONETcriteria\\_final1\\_excerpt.pdf](http://aeronet.gsfc.nasa.gov/new_web/Documents/AERONETcriteria_final1_excerpt.pdf)).

127 However, a less restrictive threshold is applied to the AOD, which we restricted to cases with  
128  $AOD_{440nm} > 0.15$ , instead of 0.4. This choice must be considered a compromise between the amount  
129 and the quality of the data. This kind of approach has been adopted by other authors using  
130 AERONET absorption data (e.g. Mallet et al., 2013). The threshold of 0.15 seems adequate  
131 analyzing the typical values of the AOD in the Iberian Peninsula (e.g., Bennouna et al., 2011;  
132 Obregón et al., 2012), because it can be considered a value to separate background aerosol  
133 conditions from episodic events with moderate or high aerosol loadings. The level 1.5-filtered data  
134 of SSA and  $g$  are daily averaged in order to have one value per day. In these conditions, the  
135 estimated uncertainty of the single scattering albedo is  $\pm 0.05$ -0.07 (Dubovik et al., 2000).  
136 Furthermore, for those days presenting level 2.0 data but also measurements in the 1.5-filtered level,  
137 we tested the uncertainty of our approach. We evaluated the difference in the SSA values of the

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<sup>1</sup> 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.

153

### 154 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

161 inputs of aerosol, total ozone column (TOC), precipitable water vapor column (PWC), and surface  
162 albedo data. We performed simulations of ultraviolet (280-400 nm), visible (400-700 nm), near-  
163 infrared (700-2800 nm), and shortwave (280-2800 nm) radiation during the periods indicated in  
164 Table 1. Total ozone column is provided by the Ozone Monitoring Instrument (OMI) and Total  
165 Ozone Mapping Spectrometer (TOMS). Daily values of these instruments are obtained from the  
166 Daily Level 3 Global Gridded products, which are downloaded using the Giovanni application  
167 (<http://disc.sci.gsfc.nasa.gov/giovanni>). Level 2.0 AERONET PWC data are used in the  
168 calculations. The uncertainty of this parameter is 10-15% (Holben et al., 1998). In addition,  
169 retrievals of surface albedo at 440, 675, 870 and 1020 nm from the AERONET algorithm are also  
170 used in this work. For land surface cover, this algorithm relies on the Lie–Ross model (Lucht and  
171 Roujean, 2000), but considering the bidirectional reflectance distributions from MODIS (Moody et  
172 al., 2005).

173 Aerosol properties obtained from AERONET measurements are also used as input to the libRadtran  
174 model. Ångström coefficients,  $\alpha$  and  $\beta$ , are utilized to compute a spectral aerosol optical depth in  
175 the wavelengths of interest (Schuster et al., 2006). Ångström exponent  $\alpha$  is obtained with the  
176 measurements between 440 and 870 nm, while the turbidity  $\beta$  is obtained from the  $\alpha$  value and  
177 aerosol data at 1020 nm. Since the aerosol asymmetry factor, single scattering albedo, and surface  
178 albedo are obtained at four wavelengths from AERONET in each measurement, three different  
179 spectral regions are simulated with the libRadtran model. For computations in the UV range (280-  
180 400 nm), the AERONET retrievals of aerosol asymmetry factor, aerosol single scattering albedo,  
181 and surface albedo at 440 nm are used. The AERONET retrievals at 675 nm of the same variables  
182 are used in the visible range (400-700 nm), while in the near-infrared region (700-2800 nm) we  
183 used the average properties retrieved at 870 and 1020 nm. In each interval, these properties are  
184 considered as wavelength independent. This choice to perform the radiative transfer simulations is  
185 proven as adequate in the Appendix A. Other options in the model set-up are: extraterrestrial



186 irradiance values are taken from Gueymard (2004); profiles of temperature, air density, ozone and  
 187 other atmospheric gases are taken from the midlatitude summer/winter standard atmospheres; and  
 188 the radiative equation solver is the improved version of the discrete ordinate method of Stamnes et  
 189 al. (2000) (DISORT2) calculated by 16-streams (e.g., de Miguel et al., 2011). After computing the  
 190 solar irradiance in the different spectral intervals, the SW irradiance is evaluated by adding up the  
 191 contributions of these three spectral regions.

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

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

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

199

$$200 \quad ARE_X = (X_{aer}^{\downarrow} - X_{aer}^{\uparrow}) - (X_{NOaer}^{\downarrow} - X_{NOaer}^{\uparrow}) \quad (1)$$

201

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

204 Daily values are obtained by the integration of the hourly data during the whole day (24 h)  
 205 considering  $ARE = 0 \ Wm^{-2}$  for  $SZA > 90^\circ$  (e.g., Bush and Valero, 2003; Valenzuela et al., 2012)  
 206 and assuming cloud-free conditions along the day:

207

$$ARE_{\text{daily}} = \sum ARE_{\text{hourly}} \frac{dt}{24} \quad (2)$$

208

209 The aerosol forcing efficiency (AFE) is defined as the rate at which the radiative effect varies per  
 210 unit of AOD (e.g., Di Biagio et al., 2009; and the references therein). The linear relationship  
 211 between aerosol radiative effect and AOD is well known (see, e.g., Costa et al., 2004, 2006; Di  
 212 Biagio et al., 2009). Hence, in this study, ARE is obtained as the slope of linear fits in the ARE vs  
 213 AOD<sub>500nm</sub> relationships. Therefore, AFE values are expressed in W m<sup>-2</sup> per AOD<sub>500nm</sub>-unit (Wm<sup>-2</sup>τ<sup>-1</sup>).  
 214

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

224

#### 225 **4. Analysis of aerosol properties over the Iberian Peninsula**

226 A direct CIMEL retrieval (AOD at 440 nm) is selected to perform the climatological analysis  
 227 because the estimations of AOD<sub>500nm</sub> (used in the ARE calculations) are obtained using α values.  
 228 Hence, we minimized the impact of other uncertainty sources in the AOD analysis. Besides, the  
 229 results for AOD<sub>440nm</sub> and AOD<sub>500nm</sub> do not differ excessively. In order to identify the differences in

230 the aerosol climatology over the six sites analyzed in this study, the monthly distribution of the  
231 daily values of the  $AOD_{440nm}$  and  $\alpha$  are evaluated using the database mentioned in Table 1. All the  
232 available level 2.0 AERONET measurements are used in this section.

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

246 To go further in the characterization,  $\alpha$  allows for a better understanding of the particle size over  
247 each site. Figure 2 shows the climatology of this variable over the six stations using also box  
248 whisker plots. Analyzing the monthly average means,  $\alpha$  values larger than one, indicative of the  
249 predominance of fine particles, are dominant over Barcelona, Palencia, and Évora. The other three  
250 stations (Cabo da Roca, Granada, and El Arenosillo) present monthly  $\alpha$  averages over and below 1,  
251 which means a larger variety of aerosol sizes over these stations. A seasonal dependence over  
252 Granada site is seen, with winter months dominated by fine particles (see also Lyamani et al., 2012)  
253 and summer months by larger ones (see also Navas-Guzman et al., 2013). Values of  $\alpha$  present a

254 large variability during summer which is indicative of the influence of different aerosol types  
255 including biomass burning events and Saharan dust transport (e.g., Pérez-Ramírez, 2008). The  
256 monthly distribution of  $\alpha$  is symmetric with similar average and median values through the year for  
257 the six sites.

258 With the daily AOD and  $\alpha$  values, it is possible to classify the origin of the aerosol particles.  
259 Previous studies suggest different thresholds of AOD and  $\alpha$  (e.g., Hess et al., 1998; Pace et al.,  
260 2006; Toledano et al., 2007b). A simple classification, which can be used for the whole Iberian  
261 Peninsula, of aerosol type is carried out in this study. The threshold between fine and large particles  
262 is placed at  $\alpha = 1$ , while the situations with a high aerosol load are those with  $\text{AOD}_{440\text{nm}} > 0.2$ .  
263 Therefore, aerosol particles can be classified in four types: maritime ( $\text{AOD}_{440\text{nm}} < 0.2$  and  $\alpha < 1$ ),  
264 desert dust ( $\text{AOD}_{440\text{nm}} > 0.2$  and  $\alpha < 1$ ), continental clean ( $\text{AOD}_{440\text{nm}} < 0.2$  and  $\alpha > 1$ ), and  
265 continental polluted ( $\text{AOD}_{440\text{nm}} > 0.2$  and  $\alpha > 1$ ). Note that the limit of  $\text{AOD}_{440\text{nm}} < 0.2$  is arbitrary  
266 and this value could be adjusted according to the sites, which likely produce a different distribution  
267 different distribution in the pie diagrams. Actually, even close stations can present slight differences  
268 in the  $\alpha$ -AOD classification (see, e.g., Obregón et al. 2012). However it is not the aim of this work  
269 to provide an extensive aerosol climatology, but rather to demonstrate the great variety of air  
270 masses over Iberia which transport different aerosol types. Although other types, such as biomass  
271 burning or mixed aerosols, are placed in the boundaries of these types, this simple classification can  
272 provide information about the aerosol sources for the six sites. The classification used here is line  
273 with the previous studies. For instance, Toledano et al., (2007) proposed for El Arenosillo site  
274 similar thresholds (see their Table V), although they identified continental polluted aerosols with an  
275  $\text{AOD}_{440\text{nm}}$  larger than 0.35 and  $\alpha > 1.4$ . Pace et al., (2006) proposed at Lampedusa island (Central  
276 Mediterranean) a desert dust identification when  $\text{AOD}_{440\text{nm}} \geq 0.15$  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  $AOD_{440nm}$  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  $\sim 0.2$ . 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  $AOD_{440nm}$ , 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

302 Roca) have not enough data to evaluate the yearly mean. Hence, we cannot ensure that the apparent  
303 minimum of AOD recorded that year is linked to global-scale phenomena or to more local  
304 conditions at the other sites. During 2010 a persistent negative phases of North Atlantic Oscillation  
305 (NAO) and Quasi Biennial Oscillation (QBO) indexes was observed (e.g., Steinbrecht et al., 2011),  
306 and the connection between air mass transport at global scale and particulate matter (at the surface)  
307 is proved by Pey et al., (2013) in the Eastern Iberian Peninsula.

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

319 The reasons behind the decrease in the aerosol load since the early 2000s are a mixed of  
320 anthropogenic and natural sources. As was reported by Aas et al. (2013), the particulate matter  
321 (PM) emissions in the Iberian Peninsula have decreased around 25% between 2000 and 2011.  
322 Furthermore, observational PM data in different Spanish sites have also shown a decrease trend in  
323 the 2000s (e.g., Barmpadimos et al., 2012; Cusack et al., 2012; Pey et al., 2013; Bennouna et al.,  
324 2014). This fact can be understood by the effect of the current economic crisis and the  
325 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.

334

## 335 **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:  $+4.9 (+3.2) \text{ Wm}^{-2}$  per decade in  $\text{ARE}_{\text{SW}}$ ,  $+3.3 (+2.1) \text{ Wm}^{-2}$  per decade in  $\text{ARE}_{\text{VIS}}$ ,  $+0.1 (+0.08) \text{ Wm}^{-2}$  per decade in  $\text{ARE}_{\text{NIR}}$ , and  $+0.06 (+0.03) \text{ Wm}^{-2}$  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

350 Peninsula could partially contribute to the increase in the levels of SW radiation at the surface (the  
351 brightening phenomenon) in this region reported by Sanchez-Lorenzo et al. (2013) and Mateos et al.  
352 (2013a).

353 To establish the general behavior of the ARE over the whole Iberian Peninsula, the yearly values  
354 using the six ground-based stations are evaluated. Only those years with, at least, simultaneous  
355 measurements at three sites are considered in these averages, and consequently, the time period is  
356 limited to 2004-2012. Figure 6 shows the evolution of the ARE and AOD at 500 nm for the entire  
357 peninsula. The previously discussed reduction of aerosol load in the six individual datasets is again  
358 corroborated with the mean data series. This decline produces a consequent decrease in the aerosol  
359 radiative effect at the four spectral ranges. The temporal trends of these yearly values are evaluated,  
360 and all the trends resulted statistically significant at the 95% significance level are shown in Figure  
361 6. Overall,  $ARE_{SW}$  over the Iberian Peninsula increased  $3.6 \text{ W m}^{-2}$  per decade while the aerosol  
362 reduced  $0.04 \text{ AOD}_{500\text{nm}}$ -unit per decade. The yearly aerosol radiative effects over the entire  
363 peninsula are in the ranges:  $-1.1 < ARE_{UV} < -0.7 \text{ W m}^{-2}$ ,  $-5.7 < ARE_{VIS} < -3.5 \text{ W m}^{-2}$ ,  $-2.6 <$   
364  $ARE_{NIR} < -1.6 \text{ W m}^{-2}$ , and  $-8.8 < ARE_{SW} < -5.7 \text{ W m}^{-2}$ . The larger contribution of the visible  
365 spectral region with respect to the whole solar spectrum was also noticed by Bush and Valero  
366 (2003). The relationship between ARE and  $AOD_{500\text{nm}}$  is analyzed more in detail in Section 6, when  
367 the aerosol forcing efficiency is evaluated for each ground-based station.

368 In addition to the inter-annual changes, the intra-annual behavior is also analyzed. For this purpose,  
369 the annual cycle (12 monthly means) is evaluated for the six stations (see Figure 7). A seasonal  
370 pattern is seen in  $ARE_{UV}$  and  $ARE_{VIS}$ , and therefore,  $ARE_{SW}$ . However,  $ARE_{NIR}$  does not follow a  
371 seasonal pattern, particularly at the Évora and Palencia stations given that  $ARE_{NIR}$  remains nearly  
372 constant. Small differences among the six stations are observed in the annual cycle during the cold  
373 seasons. The aerosol radiative effects are stronger during summer months. This can be related to the



374 higher likelihood of desert dust or biomass burning events over the Iberian Peninsula in these  
 375 months (e.g., Cachorro et al., 2008; Valenzuela et al., 2012), as was mentioned above. This is  
 376 corroborated by the increase of the differences among the stations during the warm season, likely  
 377 due to the variability in the impact of the desert dust episodes which strongly depend on the  
 378 geographical location of each site. The higher occurrence of large aerosol loads during the warm  
 379 seasons (see Figure 1), can explain the more negative ARE during summer and spring in Figure 7.  
 380 For instance, the Barcelona station, with the largest values of  $AOD_{440nm}$ , is the bottom curve of each  
 381 panel in Figure 7. Furthermore, the influence of mineral dust aerosol (with high aerosol optical  
 382 depth) during these months also causes strong radiative effects, as was also reported by previous  
 383 studies (e.g., Cachorro et al., 2008; Guerrero-Rascado et al., 2009; Antón et al., 2012a, 2012b;  
 384 Román et al., 2013; García et al., 2014). In addition, the bimodality of the monthly AOD  
 385 climatology mentioned in Section 4 has its impact on the radiative effects. The annual AOD cycle  
 386 (see Figure 1, El Arenosillo site) causes the inverse monthly distribution of ARE with a first  
 387 minimum in March. This effect is more clearly seen in  $ARE_{NIR}$  and  $ARE_{SW}$ .

388

## 389 **6. Aerosol radiative forcing efficiency in different spectral ranges**

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

395 To identify the influence of  $SSA$  and  $\alpha$  on AFE, this variable is calculated for several intervals of  
 396 each aerosol property. Four categories of single scattering albedo at 675 nm are established in the  
 397 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  $0.85 \geq$

398 SSA<sub>4</sub> > 0.80. Furthermore, aerosol size is classified in three intervals:  $0 \leq \alpha_1 \leq 1$ ,  $1 < \alpha_2 \leq 1.5$ , and  
 399  $1.5 < \alpha_3 \leq 2$ . Note that two intervals in the range of  $\alpha$  larger than 1 have been considered. One for  
 400 median particles and another one for fine particles, because of the relevant importance of median  
 401 size particle (continental or mixed aerosol aerosols types) over the Iberian Peninsula (see Figure 3).  
 402 Although the general classification between fine and coarse particles requires a more refined  
 403 classification (Schuster et al., 2006; Prats et al., 2011), the more general intervals selected in this  
 404 study are adequate to perform a study of the aerosol sizes at the six stations together.

405 Figure 8 shows the AFE obtained for the UV (AFE<sub>UV</sub>), VIS (AFE<sub>VIS</sub>), NIR (AFE<sub>NIR</sub>), and SW  
 406 (AFE<sub>SW</sub>) ranges for all these intervals. The threshold to evaluate the average in each sub-interval is  
 407 fixed at 10 data points. From these figures it is seen that, the stronger the absorption by aerosols, the  
 408 stronger their forcing efficiency. That is a decrease in the absolute values of the AFE for increasing  
 409 SSA and for all particle size. In general, the groups of non-absorbing particles exhibit a good  
 410 agreement among the six stations (see, for instance, AFE values in all the spectral ranges in the  
 411 interval  $1 < \alpha \leq 1.5$ ). Larger differences are obtained in the case of more absorbing aerosol particles.  
 412 These can be understood because of the different types of aerosols presented over each site (see  
 413 Section 4) and the different data numbers. The average AFE values over the whole Iberian  
 414 Peninsula (considering the six stations together) are presented in Table 3 as a function of  $\alpha$  and  
 415 SSA, separately. The role played by the aerosol size on AFE values is different in the three sub-  
 416 intervals of the shortwave radiation. AFE<sub>UV</sub> and AFE<sub>VIS</sub> are larger (in absolute value) for fine  
 417 particles, while the opposite occurs in the case of AFE<sub>NIR</sub>. As a result of these mixed effects, AFE<sub>SW</sub>  
 418 shows also a decrease in its values with increasing  $\alpha$ , but this effect is weaker than for the visible  
 419 and ultraviolet parts. SSA exhibits a more dominant role. As was observed before, the most  
 420 negative values are achieved for the most absorbing aerosols considered in this study (group 1 of  
 421 SSA, see Table 3).

422 The average values of forcing efficiency obtained in this study (see Table 3) are in line with those  
423 found by other authors. Table 4 summarizes the results obtained by previous studies. It is difficult to  
424 assess some features in the comparison with previous reported AFE values, because of the different  
425 aerosol types, time periods and methods that are analyzed. Our study presents the evaluation of  
426 ARE with six long-term databases of aerosol properties. In spite of that, the values shown in Table  
427 3 agree with those of Table 4, but the larger discrepancies are observed with the studies focusing on  
428 specific events. Our results match better with the results reported by, e.g., Zhou et al. (2005),  
429 Meloni et al. (2005), and Di Biagio et al. (2010). As was noticed by, e.g., Costa et al. (2004, 2006)  
430 and Di Biagio et al. (2010), AFE at the surface is larger (in absolute term) for aerosols characterized  
431 by smaller and absorbing particles. This result is corroborated by the findings shown in this study.  
432 Furthermore, as was pointed out by Di Biagio et al. (2010), the aerosol absorption is the dominant  
433 factor on AFE evaluated at the surface.

434 To evaluate the contribution of each spectral range with respect to the shortwave, the dependence of  
435 each AFE ratio (VIS to SW and NIR to SW) on SSA and  $\alpha$  is shown in Figure 9.  $AFE_{VIS}/AFE_{SW}$   
436 and  $AFE_{NIR}/AFE_{SW}$  ratios are shown in the figure since their contributions are the dominant.  
437  $AFE_{UV}/AFE_{SW}$  ratio can be obtained as 100% minus the sum of the percentage of the two other  
438 ranges. As expected, non substantial differences are observed in the behavior of the six stations  
439 considered in this study. The NIR contribution becomes more decisive for large particles ( $\alpha < 1$ ). It  
440 is expected that larger particles interact more with the longer wavelengths, while the smaller  
441 particles present more interaction with the shorter wavelengths. The presence of large particles with  
442 low SSA (high absorption) leads to a reduction of the  $AFE_{NIR}/AFE_{SW}$  ratio as well as an increase of  
443 the  $AFE_{VIS}/AFE_{SW}$  ratio. However, for non-absorbing (high SSA) large particles, the  
444  $AFE_{NIR}/AFE_{SW}$  ratio increases, and the contributions of the visible and infrared parts become more  
445 similar (both around ~40-50%). The difference between  $AFE_{VIS}/AFE_{SW}$  and  $AFE_{NIR}/AFE_{SW}$   
446 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  $AFE_{VIS}/AFE_{SW}$  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  $AOD_{500nm}$  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<sub>440nm</sub>-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<sub>UV</sub> ranges between -1.1 and -0.7 Wm<sup>-2</sup>, ARE<sub>VIS</sub> ranges between -5.7 and -3.6 Wm<sup>-2</sup>, and ARE<sub>NIR</sub> has values between -2.6 and -1.6 Wm<sup>-2</sup>. As a result, ARE<sub>SW</sub> is in the range between -8.8 and -5.7 Wm<sup>-2</sup>. The temporal trends of ARE<sub>UV</sub>, ARE<sub>VIS</sub>, ARE<sub>NIR</sub>, and ARE<sub>SW</sub> exhibit positive statistically significant trends between 2004 and 2012. For instance, the trend rate for the ARE<sub>SW</sub> is +3.6 Wm<sup>-2</sup> 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<sub>UV</sub> = -6 Wm<sup>-2</sup> $\tau^{-1}$ , AFE<sub>VIS</sub> = -34 Wm<sup>-2</sup> $\tau^{-1}$ , AFE<sub>NIR</sub> = -19 Wm<sup>-2</sup> $\tau^{-1}$ , and AFE<sub>SW</sub> = -59 Wm<sup>-2</sup> $\tau^{-1}$ .

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.

## 493 **Appendix A**

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

496 First at all, as it is mentioned in the text, most of the data present  $AOD_{440nm} < 0.15$  (~70% for  
497 Palencia, Granada, and Évora sites). For these low values,  $SSA = 0.9$  and  $g = 0.75$  are selected by  
498 the representativeness of the local aerosols in the six sites of study. To analyze possible  
499 uncertainties emerging from this choice, Table A1 shows the relative difference between two  
500 extreme cases of SSA ( $SSA_1 = 0.8$  and  $SSA_2 = 1.0$ ) for different values of the asymmetry factor and  
501 two different SZAs ( $30^\circ$  and  $60^\circ$ ). The two optical properties are also fixed as non-wavelength-  
502 dependent in this analysis. The  $AOD_{440nm}$  used is 0.15, the worst scenario possible for these cases  
503 because the higher the AOD the stronger the impact of aerosol properties. The simulations are  
504 performed for the four spectral ranges, and the net flux at the surface is evaluated for all of them.  
505 The change of SSA between the two extreme cases produce a relative difference in the SW range  
506 less than 5%, the relative difference is higher for the UV range but also  $< 10\%$ . Furthermore, as the  
507 SSA influences the diffuse radiation, the worst results are obtained at large SZAs. The impact of  $g$   
508 on the net fluxes is very weak. In conclusion, the choice of  $SSA = 0.9$  and  $g = 0.75$  in a clean  
509 scenario ( $AOD_{440nm} < 0.15$ ) is proven as adequate because of two reasons: a) representativeness of  
510 the local aerosols which can be mixture of different types, and b) the low uncertainty produced in  
511 the simulations by SSA and  $g$  under these conditions.

512 The choice of fixed SSA and  $g$  values within each of the spectral ranges (UV, VIS, and NIR)  
513 represented by the CIMEL spectral measurements is also justified here. The aerosol models by  
514 Shettle (1989) included in the libRadtran code (see Mayer and Kylling, 2005) are used to evaluate  
515 the uncertainty of using this approximation. The continental clean aerosols (most common type in  
516 the Iberian Peninsula, see Figure 3), and continental polluted aerosols (also very common, which

517 present an extreme case of absorption) are tested in this analysis. The simulations are performed for  
 518 the expected spectral behavior of SSA and  $g$  following Shettle (1989) and the case of fixed  
 519 properties in the UV (SSA and  $g$  values at 440 nm), VIS (SSA and  $g$  values at 675 nm), and NIR  
 520 intervals (SSA and  $g$  average of values at 870 and 1020 nm). Figure A1 presents the evolution of  
 521 the relative error (considering as reference the net flux with the expected spectral dependence of  
 522 aerosol properties) for several AOD values between aerosol-free and  $\text{AOD}_{550\text{nm}} = 0.6$ . In the case of  
 523 continental clean aerosols (Figure A1.a), the error of using our assumption is lower than 5% for all  
 524 SZAs and spectral ranges. Therefore, as the large majority of aerosol particles are of this type, the  
 525 methodology used and proposed in this study only introduces a relative error below 5% in the  
 526 majority of the simulations. With respect to the continental polluted aerosols (Figure A1.b), the  
 527 error increases achieving a maximum around 20% for the UV range and very turbid conditions. For  
 528 large AOD conditions in the Iberian Peninsula (e.g.,  $\text{AOD}_{550\text{nm}} = 0.4$ ) but with low frequency of  
 529 occurrence in contrast to  $\text{AOD}_{440\text{nm}} < 0.15$ , the error of the SW range is below 5%. However, the  
 530 UV range is more sensitive to our method and the error is around 15% at  $\text{SZA} = 60^\circ$ . As it was  
 531 mentioned above, the errors are larger for large SZAs because of the possible interaction between  
 532 absorption and scattering processes resulting the diffuse radiation. The visible range is more  
 533 sensitive to the spectral variations than the NIR interval, which exhibits a maximum error around  
 534 11% at  $\text{SZA} = 60^\circ$  and  $\text{AOD}_{550\text{nm}} = 0.6$ . The daily net radiative fluxes are also evaluated for the two  
 535 aerosol types in order to quantify the uncertainty in the final simulated data used in this study. For  
 536 Palencia site (and the corresponding SZA evolution), a daily value for the June 20th is simulated  
 537 assuming  $\text{TOC} = 300$  DU and  $\text{PWC} = 1$  cm. The results for the continental polluted case with  
 538  $\text{AOD}_{440\text{nm}} = 0.4$  exhibit differences between the spectral and fixed-band aerosol properties of: 7.5%,  
 539 5.3%, 4.0%, and 4.8% for the UV, VIS, NIR, and SW intervals. The relative errors for the same  
 540 intervals with continental clean (and same AOD value) are: 1.9%, 1.2%, 1.4%, and 1.4%,  
 541 respectively. Therefore, the uncertainty due to fixed optical properties in each spectral range is

542 dependent on the aerosol type but the error caused can be considered as acceptable. Since actual  
543 aerosols often present mixtures of different types, the uncertainty of using the theoretical spectral  
544 evolution for one type (given by an aerosol model) can also produce uncertainties which should be  
545 taken into account. Although other aerosol types are not tested in this analysis, a similar behavior  
546 can be expected. For instance, for the case of desert dust aerosols, Román et al. (2013) found a  
547 slight influence of spectral aerosol absorption properties on UV irradiance analyzing a strong  
548 Saharan intrusion over Granada site.

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

553

554

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572

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844

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

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

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

	AERONET database	Estimated uncertainty
AOD	Level 2.0	$\pm 0.01$ -0.02
SSA, g (AOD <sub>440</sub> >0.4)	Level 2.0	$\pm 0.03$ (in SSA)
SSA, g (0.15<AOD <sub>440</sub> <0.4)	Level 1.5-filtered*	$\pm 0.05$ -0.07(in SSA)
SSA, g (AOD <sub>440</sub> <0.15)	Fixed value	
PWC	Level 2.0	10-15%

856 \*Filters applied are the same as in level 2.0 except for AOD<sub>440</sub> (see text).

861 **Table 3.** AFE values and their standard error for the UV, VIS, NIR, and SW ranges for, separately,  
862 four SSA and three  $\alpha$  intervals over the Iberian Peninsula. Units are  $\text{Wm}^{-2}\tau^{-1}$ . SSA groups:  $0.85 \geq$   
863  $\text{SSA}_1 > 0.80$  (group 1),  $0.90 \geq \text{SSA}_2 > 0.85$  (group 2),  $0.95 \geq \text{SSA}_3 > 0.90$  (group 3), and  $1.0 \geq$   
864  $\text{SSA}_4 > 0.95$  (group 4); and  $\alpha$  groups:  $0 \leq \alpha_1 \leq 1$  (group 1),  $1.0 \leq \alpha_2 \leq 1.5$  (group 2), and  $1.5 < \alpha_3 \leq 2$   
865 (group 3). The average values without any classification are also presented.

Variable	Group	AFE <sub>UV</sub>	AFE <sub>VIS</sub>	AFE <sub>NIR</sub>	AFE <sub>SW</sub>
$\alpha$	1	$-5.41 \pm 0.06$	$-30.1 \pm 0.3$	$-20.9 \pm 0.2$	$-56.5 \pm 0.5$
	2	$-6.60 \pm 0.09$	$-38.3 \pm 0.4$	$-19.1 \pm 0.2$	$-64.0 \pm 0.6$
	3	$-7.06 \pm 0.10$	$-39.4 \pm 0.4$	$-16.9 \pm 0.2$	$-63.3 \pm 0.7$
SSA	1	$-9.7 \pm 0.2$	$-52.8 \pm 0.8$	$-24.9 \pm 0.5$	$-87.4 \pm 1.4$
	2	$-8.19 \pm 0.10$	$-44.6 \pm 0.4$	$-21.2 \pm 0.2$	$-74.0 \pm 0.6$
	3	$-6.37 \pm 0.05$	$-35.9 \pm 0.2$	$-19.5 \pm 0.2$	$-61.8 \pm 0.3$
	4	$-4.59 \pm 0.05$	$-26.6 \pm 0.2$	$-18.1 \pm 0.2$	$-49.3 \pm 0.3$
Average		$-5.98 \pm 0.05$	$-33.7 \pm 0.2$	$-19.34 \pm 0.11$	$-59.1 \pm 0.3$

**Table 4.** Daily Forcing Efficiencies at the surface by previous studies. Legend: desert dust (DD), continental-biomass burning (C-BB), and forest fires (FF).

Reference	Aerosol Type	AFE <sub>x</sub>	Value (Wm <sup>-2</sup> τ <sup>-1</sup> )	Time period	Region	More info.
Díaz et al. (2007)	Mixed	AFE <sub>UV</sub>	-3	July 2002	Spain	290-363 nm
Meloni et al. (2005)	DD Mixed	AFE <sub>VIS</sub>	-28.4 -45.6	July 2002	Central Mediterranean	
Lyamani et al. (2006)	Mixed	AFE <sub>VIS</sub>	-75.8	August 2003	Spain	2003 heat wave
Di Biagio et al. (2010)	DD C-BB Mixed	AFE <sub>SW</sub>	-68.9 -59.0 -94.9	2004-2007	Central Mediterranean	At the equinox
Esteve et al. (2014)	Mixed	AFE <sub>SW</sub>	-139.0	2003-2011	Spain	200 cloud-free days
Santos et al. (2008)	FF	AFE <sub>SW</sub>	-113.0	2004-2005	Portugal	Absorbing aerosols
di Sarra et al. (2011)	DD	AFE <sub>SW</sub>	-55	25-26/03/2010	Central Mediterranean	Strong event
García et al. (2014)	DD	AFE <sub>SW</sub>	-59	2009-2012	Canary Islands	386 cloud-free days
Costa et al. (2006)	DD	AFE <sub>SW</sub>	-116.9	7/04/2000	Korea	SSA = 0.76
Zhou et al. (2005)	DD	AFE <sub>SW</sub>	-80/-48	Monthly aerosol climatology	North Africa and Arabian Peninsula	Depending on surface albedo
Saha et al. (2008)	C-BB Mixed	AFE <sub>SW</sub>	-97.6 -81.5	2005-2006	French Mediterranean	0.7 < SSA < 0.8
Valenzuela et al. (2012)	DD	AFE <sub>SW</sub>	-70	2005-2010	Spain	

**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.

SZA	g	RD <sub>UV</sub> (%)	RD <sub>VIS</sub> (%)	RD <sub>NIR</sub> (%)	RD <sub>SW</sub> (%)
30°	0.65	-6.5	-3.7	-1.8	-3.0
	0.7	-6.5	-3.7	-1.8	-3.0
	0.75	-6.6	-3.8	-1.9	-3.1
	0.8	-6.7	-3.8	-1.9	-3.1
60°	0.65	-9.3	-5.7	-2.8	-4.5
	0.7	-9.4	-5.8	-2.9	-4.6
	0.75	-9.5	-5.9	-3.0	-4.7
	0.8	-9.6	-6.0	-3.0	-4.8

880 Figure Captions

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

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

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

887 **Figure 4.** Yearly values of AOD<sub>440nm</sub> at the six sites: Barcelona (blue diamonds), Palencia (purple  
888 triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El  
889 Arenosillo (green circles). The text points out the statistically significant trend obtained.

890 **Figure 5.** Evolution of yearly ARE<sub>UV</sub> (a), ARE<sub>VIS</sub> (b), ARE<sub>NIR</sub> (c), and ARE<sub>SW</sub> (d) at the six sites:  
891 Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey  
892 crosses), Granada (black stars), and El Arenosillo (green circles). Vertical bars indicate the standard  
893 deviation of each yearly value at Palencia station.

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

898 **Figure 7.** Annual cycle of ARE<sub>UV</sub> (a), ARE<sub>VIS</sub> (b), ARE<sub>NIR</sub> (c), and ARE<sub>SW</sub> (d) at the six sites:  
899 Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey  
900 crosses), Granada (black stars), and El Arenosillo (green circles). Vertical bars point out the  
901 standard deviation of each monthly value at Évora station.

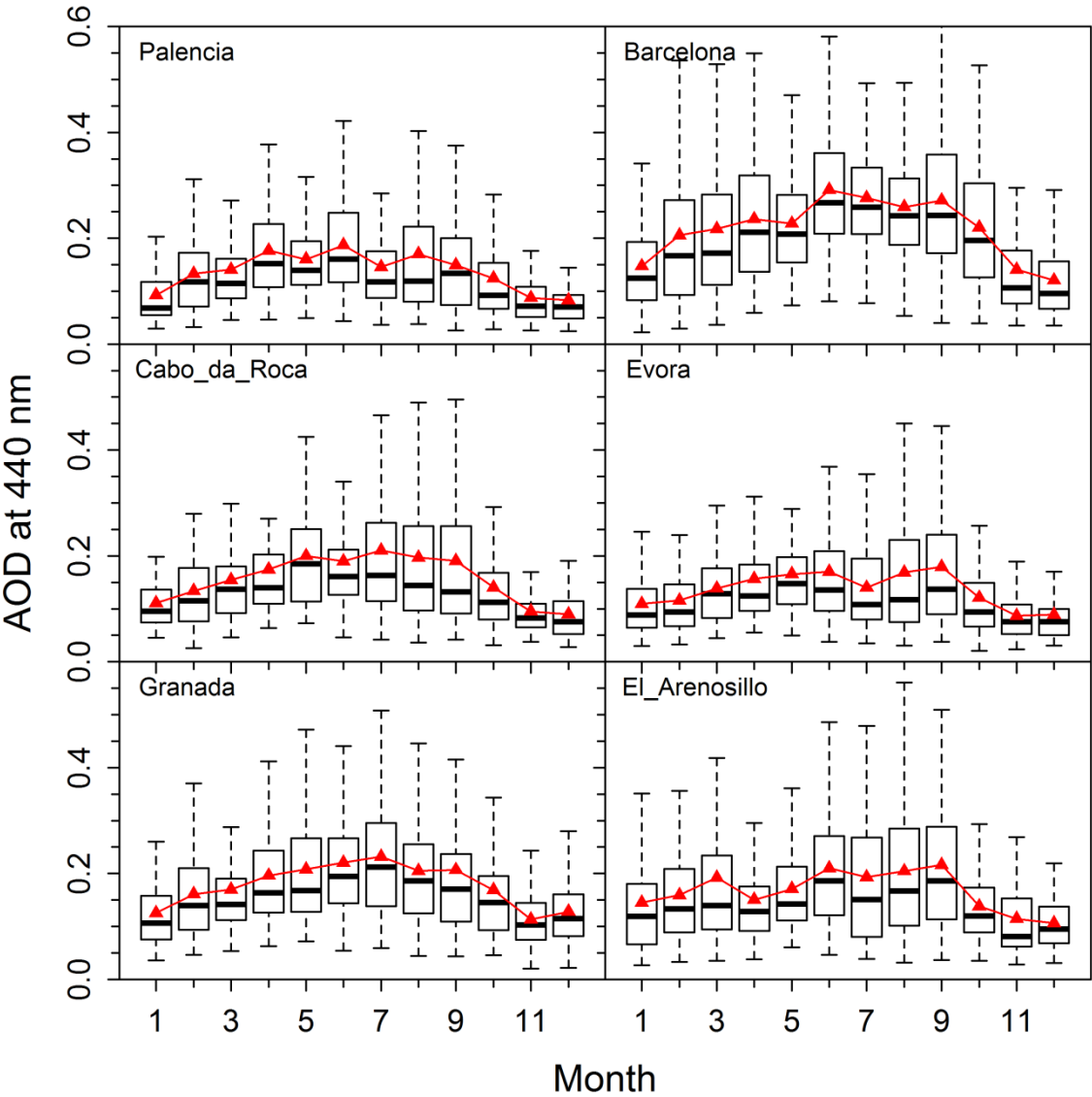


902 **Figure 8.**  $AFE_{UV}$ ,  $AFE_{VIS}$ ,  $AFE_{NIR}$ , and  $AFE_{SW}$  against four groups of aerosol single scattering  
903 albedo and three intervals of  $\alpha$  at the six sites: Barcelona (blue diamonds), Palencia (purple  
904 triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El  
905 Arenosillo (green circles).

906 **Figure 9.** Dependence of  $AFE_{VIS}/AFE_{SW}$  (a, c, e) and  $AFE_{NIR}/AFE_{SW}$  (b, d, f) ratios on SSA for  
907 large (a, b), medium (c, d) and small (e, f) particles at the six sites: Barcelona (blue diamonds),  
908 Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars),  
909 and El Arenosillo (green circles).

910

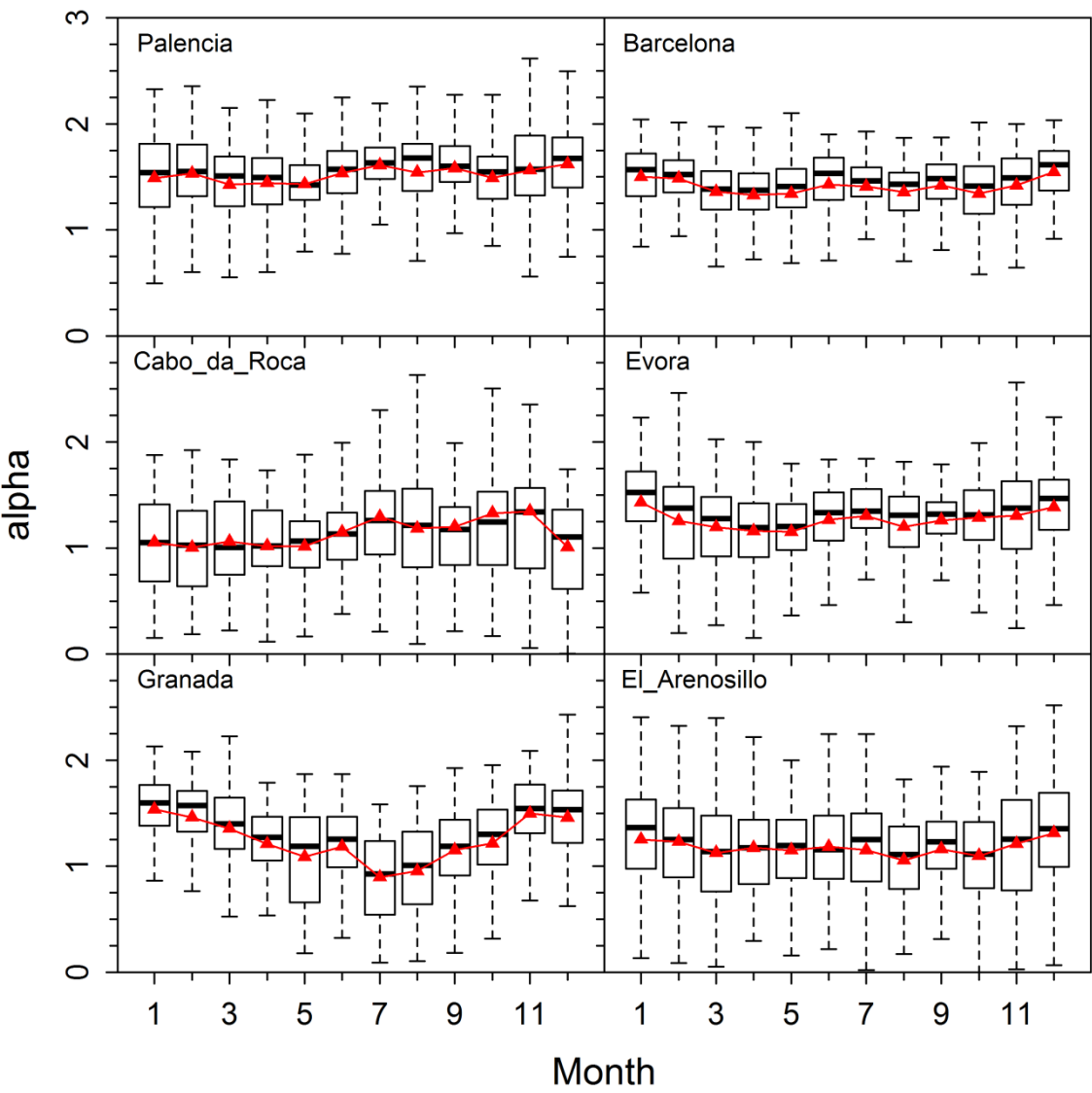
911    Figure 1



912

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

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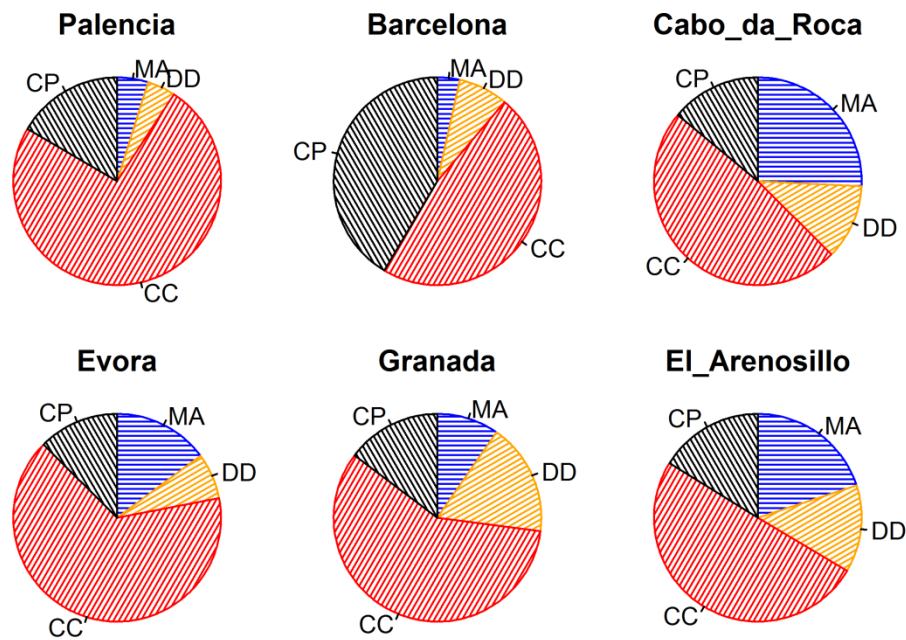
918 **Figure 2.** Annual cycle of daily values of  $\alpha$  ('alpha' in the figure) by box whisker plots. Triangles  
919 and horizontal solid lines indicate the monthly average and median values, respectively.

920

921

922 Figure 3

923



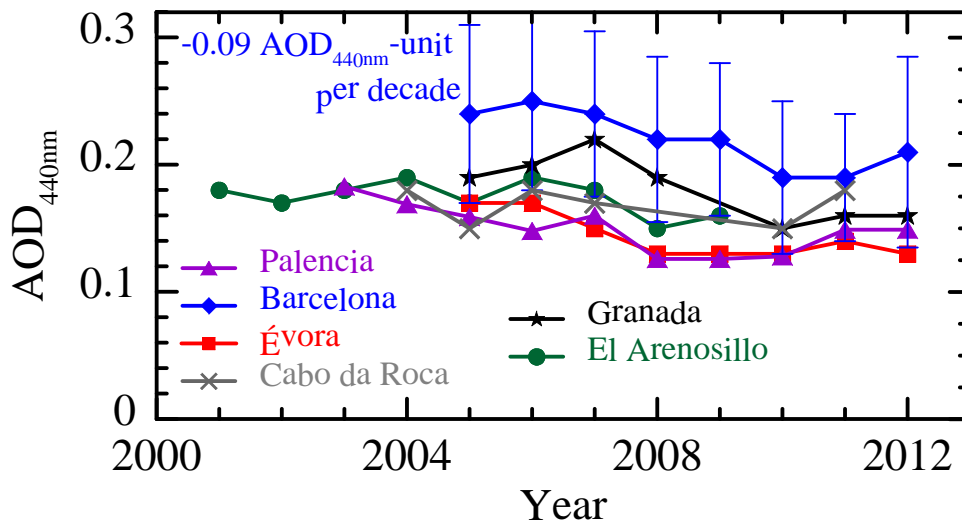
924

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

927

928 Figure 4

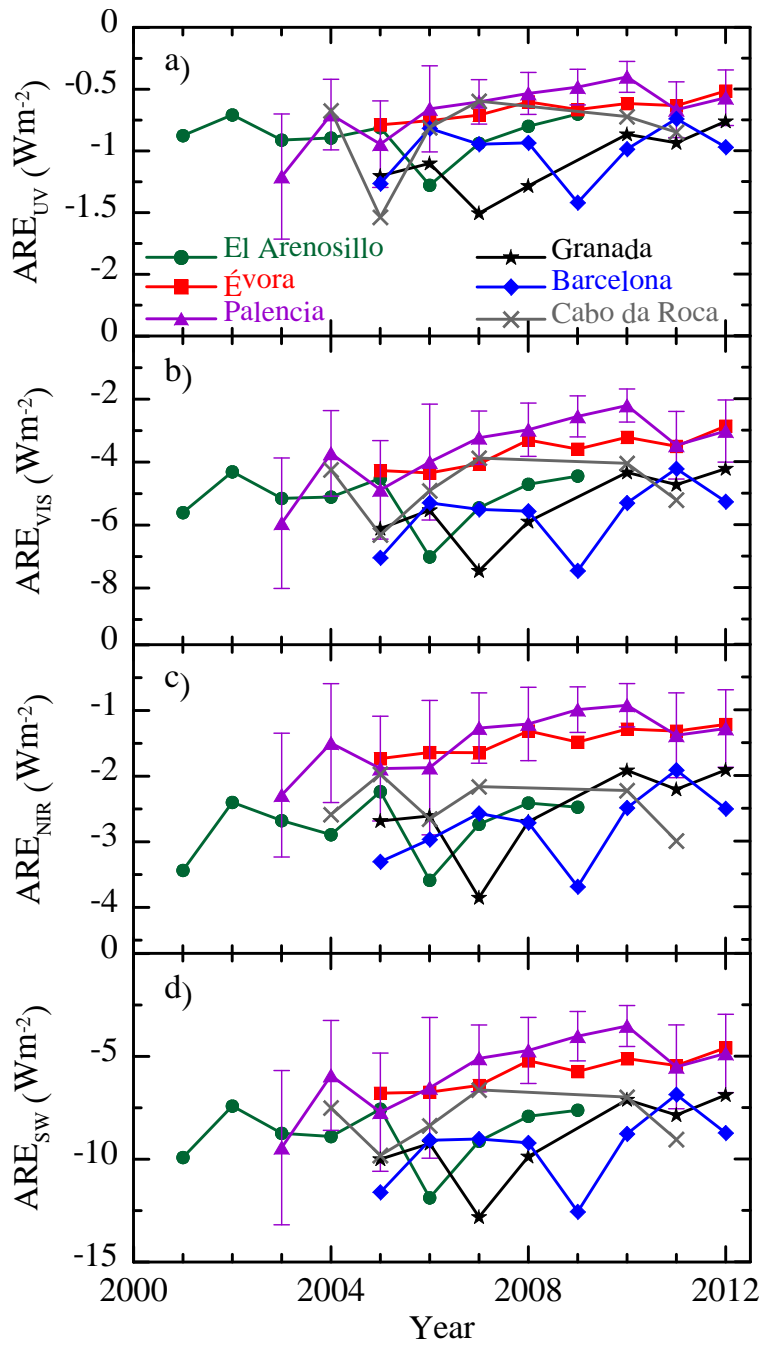
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930

931 **Figure 4.** Yearly values of AOD<sub>440nm</sub> at the six sites: Barcelona (blue diamonds), Palencia (purple  
932 triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El  
933 Arenosillo (green circles). The text points out the statistically significant trend obtained. Vertical  
934 bars indicate the standard deviation of each yearly value at Barcelona station.

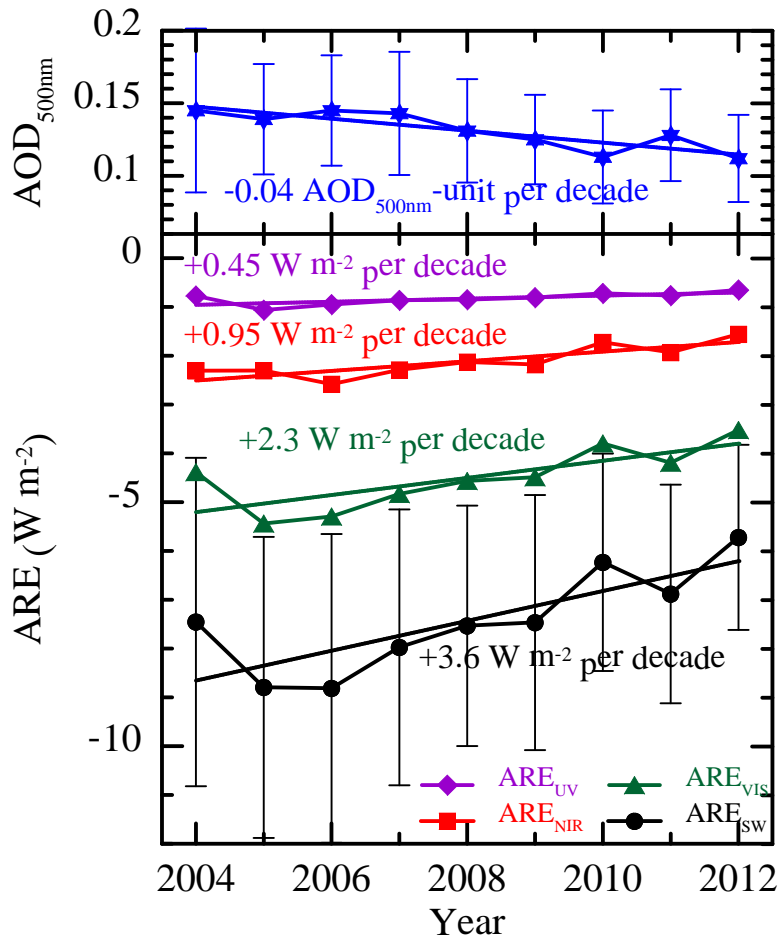
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937

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

942 Figure 6  
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944

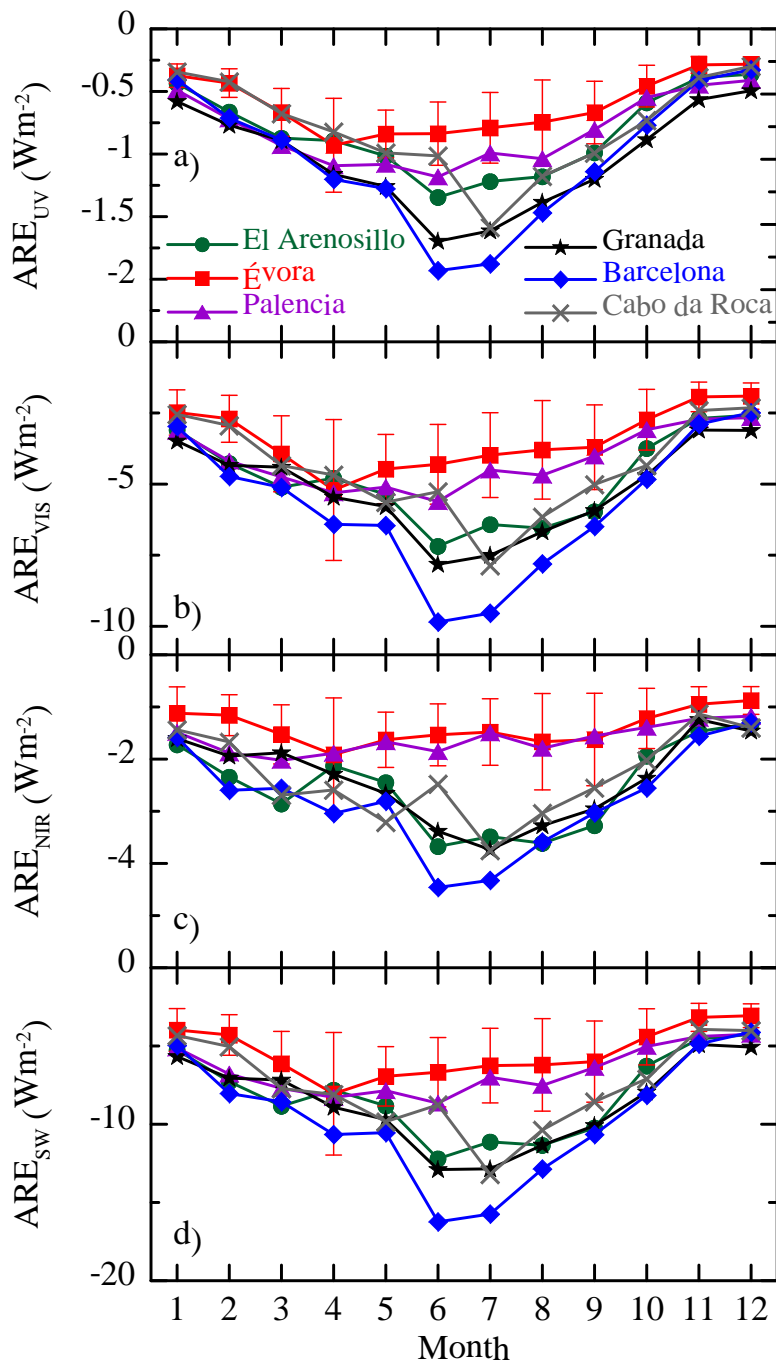


945  
946

947 **Figure 6.** Evolution of annual ARE at the four spectral ranges (ARE<sub>UV</sub> purple diamonds, ARE<sub>VIS</sub>  
948 red squares, ARE<sub>NIR</sub> green triangles, and ARE<sub>SW</sub> black circles) and AOD at 500 nm (blue stars)  
949 averaging the data from the six Iberian ground-based sites (only years with, at least, three sites are  
950 considered). Dashed lines point out the linear trends (see text). Vertical bars indicate the standard  
951 deviation.

952

953 Figure 7  
954

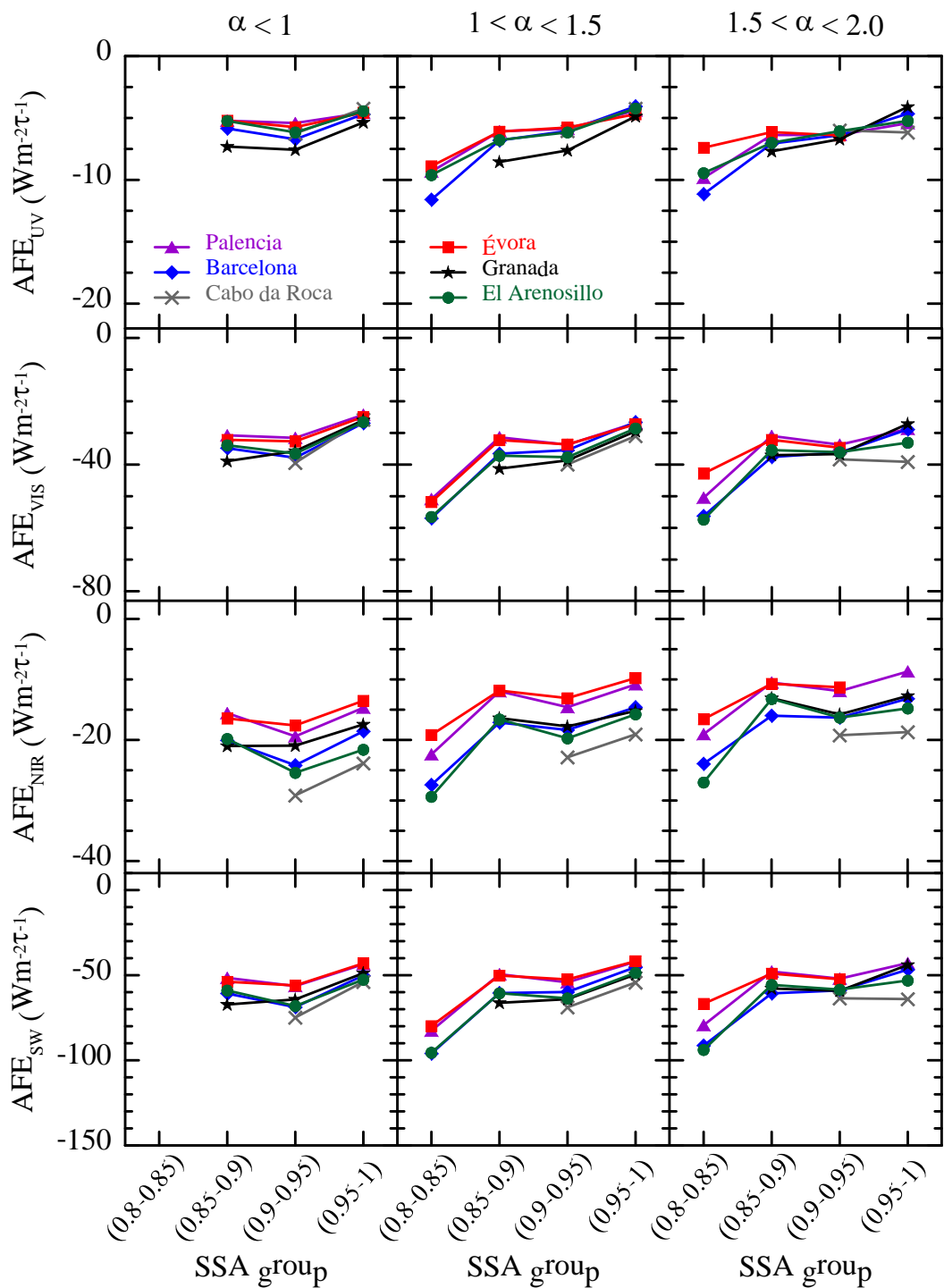


955

956 **Figure 7.** Annual cycle of  $\text{ARE}_{\text{UV}}$  (a),  $\text{ARE}_{\text{VIS}}$  (b),  $\text{ARE}_{\text{NIR}}$  (c), and  $\text{ARE}_{\text{SW}}$  (d) at the six sites:  
957 Barcelona (blue diamonds), Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey  
958 crosses), Granada (black stars), and El Arenosillo (green circles). Vertical bars point out the  
959 standard deviation of each monthly value at Évora station.



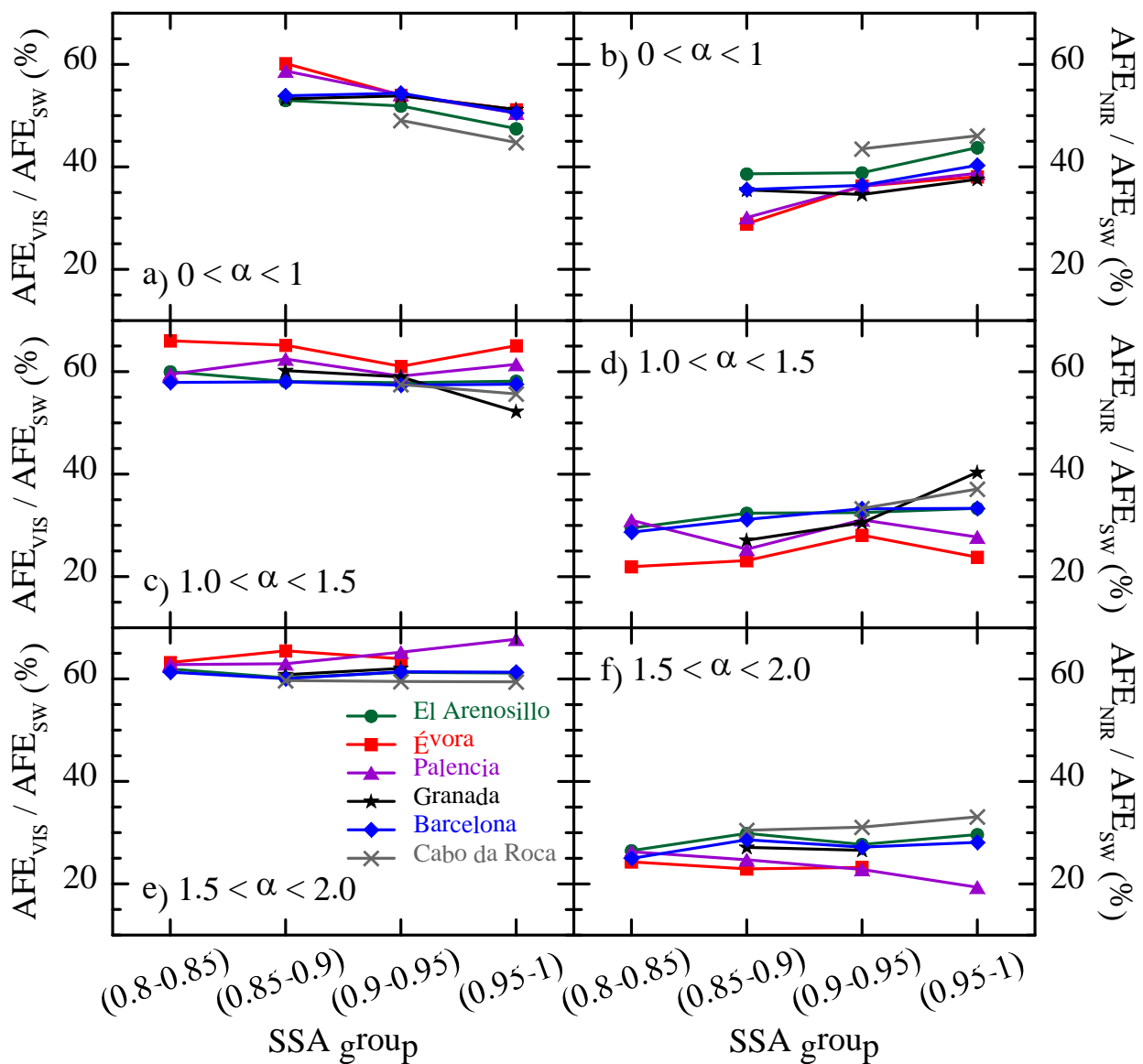
960 Figure 8



961

962 **Figure 8.**  $AFE_{UV}$ ,  $AFE_{VIS}$ ,  $AFE_{NIR}$ , and  $AFE_{SW}$  against four groups of aerosol single scattering  
963 albedo and three intervals of  $\alpha$  at the six sites: Barcelona (blue diamonds), Palencia (purple  
964 triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars), and El  
965 Arenosillo (green circles).

966 Figure 9  
967

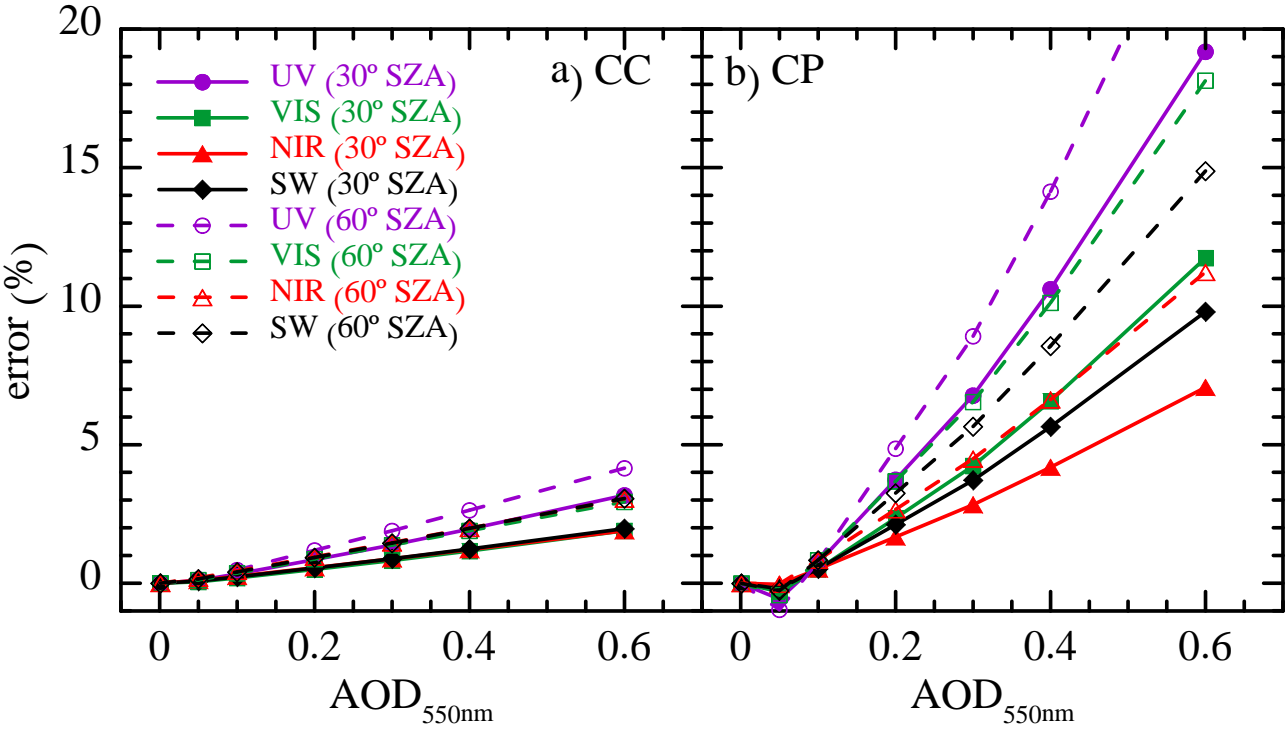


968  
969 **Figure 9.** Dependence of  $AFE_{VIS}/AFE_{SW}$  (a, c, e) and  $AFE_{NIR}/AFE_{SW}$  (b, d, f) ratios on SSA for  
970 large (a, b), medium (c, d) and small (e, f) particles at the six sites: Barcelona (blue diamonds),  
971 Palencia (purple triangles), Évora (red squares), Cabo da Roca (grey crosses), Granada (black stars),  
972 and El Arenosillo (green circles).

973

974 Figure A1

975



976

977 Figure A1. Evolution of the error committed when the optical properties are fixed in the different  
978 spectral range for two SZAs, and continental clean (CC, a) and continental polluted (CP, b)  
979 aerosols.

980