

Aerosol optical, microphysical and radiative forcing properties during variable intensity African dust events in the Iberian Peninsula



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ABSTRACT

Aerosol measurements at two AERONET (AErosol RObotic NETwork) sites of the Iberian Peninsula: Madrid (40°.45N, 3.72W) and La Coruña (43°.36N, 8°.42W) have been analyzed for the period 2012–2015 to assess aerosol optical properties (intensive and extensive) throughout the atmospheric column and their radiative forcing (RF) and radiative forcing efficiency (RF^{eff}) estimates at the Bottom and Top Of Atmosphere (BOA and TOA respectively). Specific conditions as dust-free and African dust have been considered for the study. Unprecedented, this work uses the quantification of the African dust aerosol at ground level which allows us to study such AERONET products at different intensity levels of African events: Low (L), High (H) and very high (VH). The statistical difference between dust-free and African dust conditions on the aforementioned parameters, quantified by means of the non-parametric Kolmogorov-Smirnov test, is quite clear in Madrid, however it is not in La Coruña. Scattering Angstrom Exponent (SAE) and Absorption Angstrom Exponent (AAE) were found to be 1.64 ± 0.29 and 1.14 ± 0.23 respectively in Madrid for dust-free conditions because typical aerosol sources are traffic emissions and residential heating, and black carbon is an important compound in this aerosol kind. On the other hand, SAE and AAE were 0.96 ± 0.60 and 1.44 ± 0.51 for African dust conditions in this location. RF (at shortwave radiation) seems to decrease as the African dust contribution at ground level is larger which indicates the cooling effect of African dust aerosol in Madrid. We have also proved the potential of a 2D-cluster analysis based on AAE and SAE to differentiate both situations in Madrid. Conversely, it is suggested that aerosols observed in La Coruña under dust-free conditions might come from different sources. Then, SAE and AAE are not good enough indicators to distinguish between dust-free and African dust conditions. Besides, as La Coruña is at a further distance than Madrid from the African dust source it is believed that aerosol optical properties might significantly change due to some deposition and aging/coating process and therefore the cooling effect (RF decreases as the African dust contribution at ground level is larger) is not observed.

1. Introduction

It is known that atmospheric aerosols exert a significant influence on climate given their key role to alter the incoming solar and outgoing infrared radiation. By increasing the scattering or absorption of this radiation, aerosols can produce a negative (cooling effect) or positive (warming effect) direct radiative forcing (RF) respectively. Nowadays, this driver is quantified as -0.27 [-0.77 to 0.23] W m^{-2} due to the fact that most aerosol compounds in the atmosphere have a non-absorbing behavior (IPCC, 2013). What is more, aerosols have the potential to modify the microphysical structure, lifetime and coverage of clouds and consequently the radiative balance as well. According to the Fifth Assessment Report of the IPCC (2013), this driver, named as

“cloud adjustments due to aerosols”, presents a radiative forcing of -0.55 [-1.33 to -0.06] W m^{-2} (indirect radiative forcing).

So that, depending on aerosol optical, microphysical and chemical properties, their effects on the Earth's atmosphere radiative budget can range from negligible to very significant (Wang et al., 2009; Yu and Zhang, 2011). For this reason it is so important to accomplish a proper aerosol characterization so as to determine their radiative forcing.

However, if it were not enough, when desert dust events take place, mineral aerosols, produced by continuous soil erosion, are injected and transported throughout the atmosphere over long distances. Desert dust is chemically and physically transformed during such transport which prevents from a proper estimation of the influence of mineral aerosols on radiative forcing given that it is affected by a wide range of

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uncertainties (Sokolik and Toon, 1996). It must be taken into account that desert dust makes up about 40% of aerosol mass yearly injected into the troposphere (Andreae, 1995) and in particular Sahara desert emit half of the world atmospheric mineral dust (Prospero et al., 2002). In general, the large temporal and spatial (horizontal and vertical) variability in chemical composition and physical properties leads to a high uncertainty degree in aerosol radiative forcing estimates (Boucher et al., 2013; Forster et al., 2007) and represents one of the main difficulties to address these issues.

In Europe, ACTRIS (Aerosols Clouds and Trace gases Research Infrastructure) is currently the framework, where these matters are addressed. ACTRIS integrates different aerosol observational networks such as the European section of AERONET (Holben et al., 1998), EARLINET (European Aerosol Reseach Lidar NETwork) (Pappalardo et al., 2014) and other surface in-situ networks to provide columnar, vertically-resolved, and in-situ optical and microphysical properties respectively. Likewise, satellites as TERRA or CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) procure information from space about aerosol optical properties at different regions as they complete their orbit over the globe.

Concerning the aerosol optical properties provided by these networks, it must be noted that intensive properties are very important as they can provide inherent information about aerosols and how they behave from the radiative standpoint because they are not influenced by the aerosol burden. For instance the single scattering albedo ($SSA(\lambda)$), the ratio of scattering to extinction, is a variable frequently utilized to study the influence of aerosol on radiative forcing. By way of illustration, in-situ stations tend to combine the use of nephelometers and Particle Soot Absorption Photometers (PSAP) or aethalometers in order to estimate this latter parameter (Coz et al., 2016). Moreover, the SSA has been also used to distinguish distinct events and aerosol types (Soni et al., 2010; Yang et al., 2009). These authors estimated a SSA of 0.7 and 0.9 for urban-industrial and dust aerosols at 550 nm respectively. Nevertheless, there are other intensive aerosol optical properties that can furnish a detailed aerosol characterization. The Extinction Angstrom Exponent (EAE) can procure information about the particle size. Hence, values of EAE close to 0 point out a coarse particle predominance whereas if this parameter tends to 3 indicates that fine particles are prevalent (Costabile et al., 2013). However, several authors have indicated that EAE is not an ideal indicator to identify the average aerosol size because this parameter contains an absorption dependence. For this reason, it is convenient to study separately the Scattering Angstrom Exponent (SAE) and Absorption Angstrom Exponent (AAE) (Valenzuela et al., 2015). In this sense, pure black carbon usually presents an AAE close to 1, while on the contrary, dust particles tend to exhibit values > 2 (Bergstrom et al., 2010). Along with it, parameters as asymmetry factor or effective radius that are described throughout this paper can be very useful when it comes to this aerosol characterization.

So, this paper presents a study of aerosol optical and microphysical properties along with radiative forcing estimates at two Iberian peninsula sites: Madrid and La Coruña, observed by the AERONET stations in these locations for the 2012–2015 period. The main aim of this work is the assessment of the aerosol optical and microphysical properties under distinct atmospheric situations: desert dust and dust-free conditions (consequently aerosol optical and microphysical properties are attributed to the typical aerosol in that region) at such places in the Iberian Peninsula. Madrid and La Coruña sites were chosen to study the effect of the distance from the African dust source regions on aerosol properties in the Iberian Peninsula. Previous and similar studies of these events have been carried out in Granada (Valenzuela et al., 2015). This latter work is used in this study for comparison purposes, consequently the Iberian Peninsula is relatively well covered from this standpoint. Moreover, in principle, Madrid and La Coruña sites may offer the opportunity to study different types of aerosol under dust-free conditions (aerosol from maritime, biomass burning and traffic sources). Finally,

the relationship between these properties and aerosol radiative forcing has been evaluated.

2. Experimental sites

Aerosol optical and microphysical properties and radiative forcing estimates were obtained by means of two sun photometers, CIMEL Electronique 318-A. Both are installed at AEMET (Agencia Estatal de Meteorología, Spanish Agency of Meteorology) facilities, in Madrid (40°45'N, 3.72'W) at 680 m above sea level (asl) and in La Coruña (43.36'N, 8.42'W) at 67 m asl, both urban areas. Moreover, time series of PM_{10} daily data and African dust daily contributions were obtained from two regional background air quality monitoring sites. In particular, El Atazar station (40°.91'N, 3.46'W), which belongs to the Madrid Regional Air Quality Network, is installed at 940 m asl. This station is 58 km far from the AERONET Madrid site. On the other hand, the Noia station (42.72'N, 8.92'W), which is member of EMEP (European Monitoring and Evaluation Programme), is located at 685 m asl. Likewise, Noia station is 69 km far from the AERONET La Coruña site. They both are the closest stations to the AERONET sites respectively. Two different techniques have been used to determine PM_{10} concentrations: gravimetric determinations at the EMEP site (Noia) and real time monitors based on Beta gauge attenuation at El Atazar. In this latter site the real time concentrations were corrected against the gravimetric ones. Since only the official data reported to the European Commission are used in this work, their quality is guaranteed.

The Madrid metropolitan area is placed at the center of the Iberian Peninsula, within an air shed (the Madrid air basin) bordered to the north-northwest by a high mountain chain, Sierra de Guadarrama, 40 km from the metropolitan area, to the south by another mountain system, Montes de Toledo, and finally to the northeast and east by lower mountainous terrain. The Madrid climate is continental Mediterranean. The population of this metropolitan area and surrounding towns is nearly 6 million inhabitants, one of the most densely populated regions in Spain. The Madrid plume is considered as typically urban, fed by traffic emissions and residential heating, given that industrial activity is comprised of light factories and it does not represent an important pollutant source (Artíñano et al., 2003; Salvador et al., 2015).

On the other hand, La Coruña metropolitan area is located at the northwest edge of the Iberian Peninsula as it is a coastal city, surrounded to the north-northwest by the Atlantic ocean and bordered to the east-southeast by a mountain system, called Macizo Galaico. It is noteworthy to mention that the forestry area located in the outskirts of the metropolitan area is significant as the Comunidad Autónoma of Galicia is one of the regions in Spain which presents higher forest density estimation but also where more forest-fires occur. Concerning the population, there are approximately 250,000 inhabitants in this metropolitan area. Thus, this area is a humid and well-ventilated area frequently affected by westerly winds and frontal systems. In comparison with the typical Madrid plume, the La Coruña air basin presents less particle matter from traffic but higher contribution from other sources such as a combustion power plant and the maritime source (Salvador et al., 2007).

3. Instrumentation and methods

Measurements of columnar aerosol properties were obtained through two automatic sun and sky scanning spectral radiometers (CIMEL CE-318) deployed at Madrid and La Coruña. These instruments form part of AERONET, one of the most useful networks when it comes to atmospheric aerosols monitoring, insofar as they provide near real time observations of spectrally-resolved and column-integrated aerosol optical and microphysical properties and also because they are worldwide distributed. Furthermore, AERONET is in charge of calibration, processing and standardization of these instruments (Holben et al.,

1998).

These CIMEL radiometers hold a sensor head fitted with 25 cm collimators attached to a 40 cm robot base that points the sensor head either at the sun or sky within several programmed sequences. These instruments are utterly described in Holben et al. (1998). Direct sun measurements are performed with a 1.2° full field of view at 340, 380, 440, 500, 670, 870, 940 and 1020 nm. To scan all 8 wavelengths takes approximately 8 s (repeated three times within a minute). For that, 8 interference filters at the aforementioned wavelengths are installed in a filter wheel that is rotated by a direct drive stepping motor. These solar extinction measurements are then used to infer the aerosol optical depth (AOD) at each wavelength other than at 940 nm, which is the channel used to retrieve total column water vapour. With regard to sky radiance measurements (almucantar configuration), they are performed in four spectral bands: 440, 670, 870 and 1020 nm and provide aerosol microphysical parameters such as size distribution, refractive index or single scattering albedo through the inversion algorithm developed by Dubovik and King (2000) and Dubovik et al. (2006).

AERONET data were downloaded from (<http://aeronet.gsfc.nasa.gov>) and subsequently aerosol microphysical and optical properties were studied. The analyzed period ranges from January 2012 to December 2015. For that, AERONET level 1.5 data were used (cloud screened data with pre and post calibrations applied) because level 2 data were very scarce for the previously mentioned period at both stations. This fact is due to the high restrictions established by AERONET as aerosol optical depth (at 440 nm) must be > 0.4 and zenith angle must be also larger than 50°. Under these conditions, the retrieval uncertainty for SSA is ± 0.03, however it is reduced to 0.02–0.07 if AOD is lower than 0.2 at 440 nm (Dubovik and King, 2000). The data format utilized for data level 1.5 is denoted as “all points” as it provides single measurements at specific date and time (when the AERONET observation was accomplished). Studies like the Valenzuela et al.'s (2015) have indicated the usefulness of AERONET level 1.5 data to characterize events like African desert dust events and its aerosols using microphysical parameters if AOD > 0.2 and solar zenith angle > 50°. So that, when the analysis of level 1.5 data was carried out, only AERONET measurements with AOD > 0.2 and solar zenith angle > 50° were used in order to analyze more accurate parameters. Daily averaged measurements provided by AERONET were not considered since this average can include single measurements with AOD < 0.2 and zenith angle < 50° and mislead to not adequate daily measurements for this study.

Hence, we have utilized the following aerosol optical and microphysical properties to characterize aerosols at both sites. AOD is the measure of aerosols allocated within a column of air from the Earth's surface to the Top Of Atmosphere. In other words, it is the degree to which aerosols prevent the transmission of light by absorption and/or scattering. The voltage (V) registered by the sun photometer is proportional to the spectral irradiance (I) reaching this instrument. The spectral irradiance estimated at the top of the atmosphere (I_0) in terms of voltage (V_0) is observed by the sun photometer deployed at Mauna Loa Observatory in Hawaii. Then, the total optical depth (τ_{tot}) is derived as follows:

$$V(\lambda) = V_0(\lambda) d^2 \exp[-\tau(\lambda)_{\text{tot}} \cdot m] \quad (1)$$

where V is the digital voltage measured at the wavelength λ , V_0 is the extraterrestrial voltage, d is the ratio of the average to the actual Earth-Sun distance and m is the optical air mass (Holben et al., 1998). Given the fact that atmospheric constituents scatter and absorb light, AOD must be estimated by subtraction such contribution from the total optical depth.

Furthermore, it is possible to make a distinction between absorption and scattering contribution exerted by aerosols over AOD using the following variables: the absorption aerosol optical depth ($\text{AOD}_{\lambda,\text{abs}}$) and scattering aerosol optical depth ($\text{AOD}_{\lambda,\text{scat}}$). They can be derived according to the following expression:

$$\text{AOD}_{\lambda,\text{abs}} = (1 - \text{SSA}_{\lambda}) \cdot \text{AOD} \quad (2)$$

$$\text{AOD}_{\lambda,\text{scat}} = \text{SSA}_{\lambda} \cdot \text{AOD} \quad (3)$$

where SSA is again the single scattering albedo. This parameter is retrieved as outlined below:

$$\text{SSA}_{\lambda} = [\sigma_{\text{scat}}(\lambda)/(\sigma_{\text{scat}}(\lambda) + \sigma_{\text{abs}}(\lambda))] \quad (4)$$

where, $\sigma_{\text{scat}}(\lambda)$, $\sigma_{\text{abs}}(\lambda)$ are the scattering and absorption coefficient. As stated before, SSA is just retrieved from sky radiance measurements at 440, 670, 870 and 1020 nm. Consequently, $\text{AOD}_{\lambda,\text{abs}}$ and $\text{AOD}_{\lambda,\text{scat}}$ are only estimated at these four wavelengths because SSA is required to carry out such estimates even though AOD is measured at a higher number of wavelengths. In addition, other intensive parameters such as Absorption Angstrom Exponent (AAE) or Scattering Angstrom Exponent (SAE) have been derived for the 440–1020 nm wavelength range as they are not estimated by AERONET. The equations are described as follows:

$$\text{AAE}(\lambda) = -\ln\left(\frac{\text{AOD}_{\text{abs}}(1020 \text{ nm})}{\text{AOD}_{\text{abs}}(440 \text{ nm})}\right) / \ln\left(\frac{1020}{440}\right) \quad (5)$$

$$\text{SAE}(\lambda) = -\ln\left(\frac{\text{AOD}_{\text{scat}}(1020 \text{ nm})}{\text{AOD}_{\text{scat}}(440 \text{ nm})}\right) / \ln\left(\frac{1020}{440}\right) \quad (6)$$

Again these parameters are useful tools to characterize aerosols. For instance, African dust or marine aerosols tend to exhibit a SAE lower than urban-industrial aerosols (Kim et al., 2005) since SAE strongly depends on size. On the other hand Russell et al. (2010) found an AAE (300–1700 nm) close to 1 for typical aerosol at US Atlantic Coast, while aerosols, affected by African dust conditions observed at Puerto Rico, showed AAE values slightly larger than 2. Other parameter utilized throughout this paper is the asymmetry factor which indicates a predominant direction of the phase function and consequently the asymmetry of the radiation scattered. Likewise, the effective radius is an area weighted mean radii of the aerosol particles.

Besides, the AERONET products have incorporated aerosol RF and aerosol RF^{eff} estimates that are inferred from microphysical parameters (size distribution, refractive index...) through a relatively new radiative transfer module. So that, solar fluxes are computed for 200 intervals throughout the 0.2 to 4 μm range by the Discrete Ordinates DISORT approach (Nakajima and Tanaka, 1988; Starnes et al., 1988). For this reason, refractive index (real and imaginary part) and surface reflectance have to be interpolated and extrapolated from the sky radiance measurement wavelengths. Finally, the GAME (Global Atmospheric Model) code (Roger et al., 2002) incorporates as inputs: the surface reflection, molecular scattering, gas absorption and aerosol scattering and absorption to retrieve aerosol radiative forcing. Further description of this code and methodology to infer such forcing from AERONET observations can be found in García et al. (2012a).

Thence, the AERONET aerosol radiative forcing is defined as the difference in global solar irradiance with and without aerosols. These parameters are estimated at two atmospheric levels: Bottom Of Atmosphere (BOA) and Top Of Atmosphere (TOA).

$$\text{RF}_{\text{BOA}} = (F_{\text{BOA}}^{\downarrow} - F_{\text{BOA}}^{\uparrow}) \cdot (1 - \text{SA}) \quad (7)$$

$$\text{RF}_{\text{TOA}} = -(F_{\text{TOA}}^{\downarrow} - F_{\text{TOA}}^{\uparrow}) \quad (8)$$

where F and F^0 stand for the broadband fluxes with and without aerosols respectively (the arrows point out the direction of fluxes, \uparrow : upward flux, and \downarrow : downward flux). SA represents surface albedo. Given that we have utilized AERONET aerosol radiative forcing estimates we have corrected RF_{BOA} by the term (1-SA) because AERONET considerers as RF_{BOA} just this difference: $(F_{\text{BOA}}^{\downarrow} - F_{\text{BOA}}^{\uparrow})$. This correction has also been carried out earlier by previous studies (García et al., 2012b) when it comes to studying these parameters provided by AERONET. Concerning RF_{TOA} , the equation coincides with AERONET radiative forcing estimates so no correction is needed in this case. Then,

we have utilized a unique value of *SA* for each almucantar retrieval, which is estimated from the spectral average of the surface albedo obtained by AERONET at 440, 670, 870 and 1020 nm.

On the other hand, since the aerosol radiative forcing highly depends on the aerosol optical depth, it seems more convenient to utilize the aerosol radiative forcing efficiency (RF^{eff}) so as to investigate which role aerosols play in the atmosphere. For this reason, aerosol radiative forcing efficiency is defined as the aerosol radiative forcing per unit of aerosol optical depth at 550 nm.

$$RF^{eff} = RF/\tau(550) \quad (9)$$

RF and RF^{eff} have been studied either at BOA and TOA. Likewise RF and RF^{eff} data under desert dust and dust-free conditions were distinguished and analyzed separately.

A robust procedure was used to identify the occurrence and duration of African dust episodic days over the central and northwestern areas of the Iberian Peninsula. It is based on the daily interpretation of air mass back trajectories computed by the HYSPLIT model (Draxler and Rolph, 2013), synoptic meteorological charts, satellite imagery (maps of aerosol index of Ozone Monitoring Instrument-OMI and NASA SeaWiFS images) and daily consultation of dust forecast models, namely: SKIRON-University of Athens (<http://forecast.uoa.gr>), BSC-DREAM8b v2.0-Barcelona Supercomputing Center (<http://www.bsc.es/projects/earthscience/DREAM/>) and NAAPS-Naval Research Laboratory (NRL), Monterey, CA (<http://www.nrlmry.navy.mil/aerosol/>). This procedure also allows quantifying the dust contribution to the PM₁₀ daily records during each potential dust episodic day by means of a statistical analysis of the time series of PM₁₀ values registered at regional background monitoring sites such as El Atazar and Noia. Hence, the higher the dust contribution levels, the more intense the dust episodic day. The feasibility of this method was demonstrated by different approaches in Escudero et al. (2007) and Viana et al. (2010). This methodology has become the Spanish and Portuguese reference method to identify and quantify African dust contributions to PM₁₀ levels since 2004. The method is also applicable across the whole Southern Europe, as demonstrated by Querol et al. (2009). Currently, this is one of the official methods recommended by the European Commission for evaluating the occurrence of African dust intrusions and quantifying its contributions (Commission staff working paper, 2011).

In this work desert dust contributions are shown as percentage to the total PM₁₀ concentrations at ground level. The African dust events were classified into three different groups: Low (L), High (H) and Very High (VH) based on African dust contribution at ground level. Therefore, L represents an African dust contribution ranging between 0 and 50%, H stands for 50–80% and VH is representative of a African dust contribution > 80%.

In addition, we have applied the Kolmogorov-Smirnov test to check the influence of the African dust in the values of the aerosol optical properties, with statistical significance. This is one of the most useful and general non-parametric method to compare two samples and it is sensitive to differences in both location and shape of the empirical cumulative distribution functions of the two samples. We have performed 6 different tests named: T₁, T₂, T₃, T₄, T₅, T₆. T₁, T₂ and T₃ stands for the comparison between dust-free against African dust conditions: L, H and VH respectively. T₄ and T₅ represent the comparison between African dust (L) against African dust (H and VH) conditions respectively. Finally T₆ stands for the likening between African dust at H and VH conditions. Then, values of $p < 0.05$ indicate statistical significant differences between the distributions at 95% confidence level.

Furthermore, we have performed a clustering analysis based on k-means method, which aims to partition the points into k groups such that the sum of squares from points to the assigned cluster centers is minimized. The iterative algorithm then repositions the centroids to the average values of the observation points in a given cluster until the

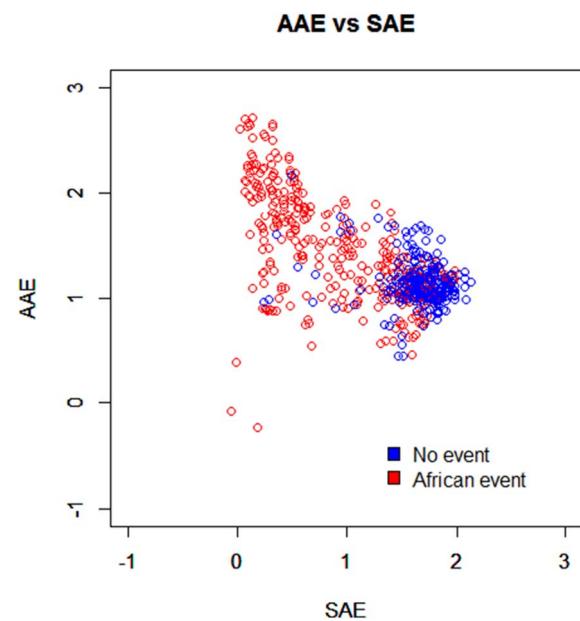


Fig. 1. Scattering and Absorption Angström exponent obtained in Madrid for African desert dust (African event) and dust-free conditions (No event) studied from AERONET 1.5 level data.

centroids remain stationary or a limit of the iteration is reached. The k-means method is based on the algorithm of Hartigan and Wong (1979). A prechosen number of k clusters (in this study k = 2, dust-free and African dust conditions), defined by the randomly seeded centroids, are populated by n observation points. The variables utilized in this cluster analysis are SAE and AAE. The purpose of this analysis is to reveal the potential of the usage of intensive optical properties (SAE and AAE) to identify certain events for example dust-free and African dust events.

4. Results and discussion

4.1. Madrid

The AERONET database products that cover from 2012 until 2015 were studied. These products were classified depending on the African dust contribution at ground level. Having considered this, intensive and extensive aerosol optical properties were analyzed to subsequently establish any relation to radiative forcing estimates when possible.

So, starting from intensive aerosol optical properties registered in Madrid must be noted the following. In Fig. 1, it is shown SAE against AAE for two different atmospheric conditions named as "No dust" and "African dust". Under dust-free conditions SAE and AAE tend to show quite constant values which in principle are attributed to the typical aerosol existent in the Madrid atmosphere. Mean values of SAE and AAE are 1.64 ± 0.29 and 1.14 ± 0.23 respectively. These values are characteristic of black carbon (in particular AAE, given that it is close to 1). As commented previously, the typical aerosol registered in Madrid is classified as urban because of the predominance of traffic emissions and residential heating (Becerril et al., 2015; Salvador et al., 2015). Conversely, aerosols observed during African dust events present more dispersed values of SAE and AAE, which is due to the mixing degree of typical Madrid aerosol with African dust aerosol (see Fig. 2). Mean values of SAE and AAE under African events are 0.96 ± 0.60 and 1.44 ± 0.51 respectively. Furthermore, the higher the African dust contribution at ground level is, the lower the SAE is and the greater the AAE is. Thus, if African dust contribution to the PM₁₀ concentration at ground level is larger than 50% (H and VH), mean values of SAE are lower than 0.5 and AAE are higher than 1.6. These results are in agreement with previous works such as Valenzuela et al.'s (2015) since they found mean values of 0.5 and 1.5 for SAE and AAE under African

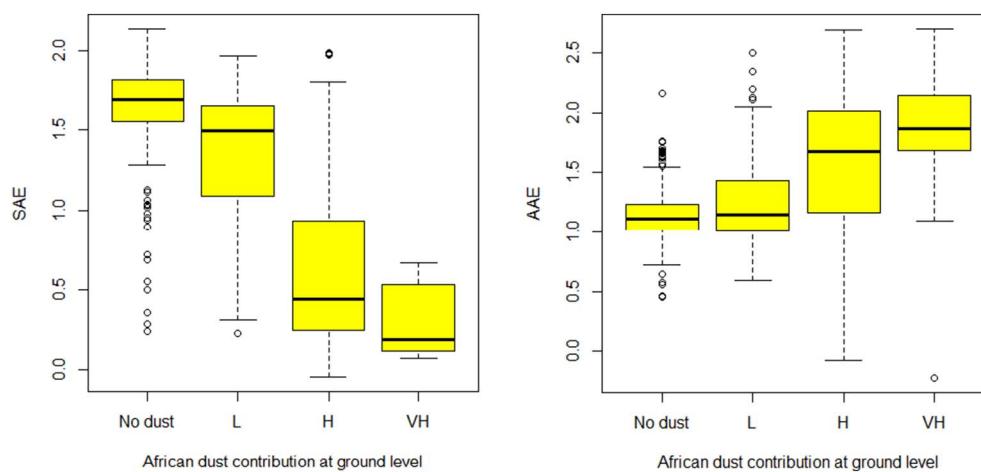


Fig. 2. Scattering and Absorption Angström exponent obtained in Madrid for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

Table 1

Summary of aerosol optical properties registered throughout the atmospheric column at different locations worldwide.

Author	Location	Aerosol type	SAE	AAE	SSA
Dubovik et al. (2002)	Solar Village, Saudi Arabia	Dust			0.92 and 0.97 (at 440 and 1020 nm)
Kim et al. (2005)	Gosan, Korea	Dust	0.38 (at 412–862 nm)		
Kim et al. (2005)	Gosan, Korea	Urban-industrial	1.3 (at 412–862 nm)		
Lyamani et al. (2006)	Granada, Spain	Urban-industrial			0.91 and 0.85 (at 440 and 1020 nm)
Perrone and Bergamo (2011)	Lecce, Italy	Dust			0.87–0.95 (at 550 nm)
Kim et al. (2011)	Tamanrasset, Algeria	Dust			0.90–0.94 (at 440 nm)
Valenzuela et al. (2015)	Granada, Spain	Dust	0.5 (at 440–1020 nm)	1.5 (at 440–1020 nm)	0.89 and 0.96 (at 440 and 1020 nm)
Valenzuela et al. (2015)	Granada, Spain	Dust-free	1.2 (at 440–1020 nm)	0.9 (at 440–1020 nm)	0.9 and 0.86 (at 440 and 1020 nm)
This study	Madrid, Spain	Dust-free	1.64 (at 440–1020 nm)	1.14 (at 440–1020 nm)	0.91 and 0.88 (at 440 and 1020 nm)
This study	Madrid, Spain	Dust	0.96 (at 440–1020 nm)	1.44 (at 440–1020 nm)	0.91 and 0.93 (at 440 and 1020 nm)
This study	La Coruña, Spain	Dust-free	1.51 (at 440–1020 nm)	1.09 (at 440–1020 nm)	0.92 and 0.88 (at 440–1020 nm)
This study	La Coruña, Spain	Dust	0.87 (at 440–1020 nm)	0.91 (at 440–1020 nm)	0.93 and 0.93 (at 440–1020 nm)

dust conditions throughout the atmospheric column explored (see Table 1).

According to these results, we state that African dust events play a relevant role on the aerosol optical properties existent in the Madrid atmosphere due to the fact that African dust aerosols behave in an opposite manner from a radiative standpoint than typical Madrid aerosols. The latter ones tend to be more absorbent whereas the first ones produce more scattering. Statistical differences are appreciated when it comes to studying SAE and AAE at African dust contribution of “No dust”, “Low”, “High” and “Very high” because p values are lower than 0.05 (see Table 2), except for the AAE comparison between cases “High” and “Very high” (column T6).

Likewise, SSA at 677, 868 and 1020 nm follow the same trend already explained (Fig. 3 left). Given that SSA is the ratio of scattering to extinction, aerosols observed under dust-free conditions revealed to have lower SSA than those aerosols existent during African dust events, because they are more absorbent and African dust aerosols produce greater scattering. This trend is slightly clearer as the wavelength is larger. However, the other way around is detected after studying SSA at 442 nm. We do not know exactly the reason why it occurs, nevertheless,

this fact has been observed in other studies (Valenzuela et al., 2015) where there is no difference between dust-free and dusty days having considered the SSA at 442 nm. In Fig. 3, it is also represented the asymmetry factor. The asymmetry factor indicates a predominant direction of the phase function and consequently the asymmetry of the radiation scattered. Therefore, values of SSA towards 1 point out that the radiation is scattered predominantly in the forward direction whereas values close to 0 indicate that radiation is scattered equally in both directions (backward and forward). There, it is observed that the higher the wavelength is, the greater difference between dusty and dust-free conditions is found when considering the asymmetry factor. What is more, aerosols observed under African dust conditions (H and VH) tend to have a greater asymmetry factor than aerosol detected in dust-free conditions or in low (L) African dust conditions. This might be due to African dust aerosols are usually greater in size which brings about a radiation scattered predominantly in the forward direction according to the Mie theory (Mie, 1908), nevertheless the asymmetry factor is also dependent on other factors that should keep in mind such as shape, etc. This statement is supported by the fact that effective radius is increased as the African dust contribution at ground level is

Table 2

The p values of the Kolmogorov-Smirnov statistical test applied to SSA, AAE, RF at BOA (efficiency), RF at BOA, RF at TOA (efficiency), RF at TOA for the Madrid atmospheric column. The p values in red stand for those that are > 0.05 and consequently no statistical difference is found.

Aerosol property	T_1	T_2	T_3	T_4	T_5	T_6
SAE	6,78E – 14	0	9,99E – 16	0	6,83E – 13	1,26E – 02
AAE	8,01E – 03	0	8,43E – 11	6,98E – 12	1,57E – 07	8,60E – 02
RF at BOA (efficiency)	1,78E – 01	2,95E – 06	1,52E – 02	3,21E – 04	1,01E – 02	3,94E – 01
RF at BOA	9,63E – 04	3,19E – 13	1,18E – 04	1,56E – 05	3,63E – 04	1,18E – 09
RF at TOA (efficiency)	5,75E – 02	2,10E – 02	8,51E – 03	7,56E – 01	1,25E – 02	2,11E – 02
RF at TOA	9,31E – 03	1,33E – 15	2,92E – 03	2,03E – 07	5,28E – 02	6,95E – 01

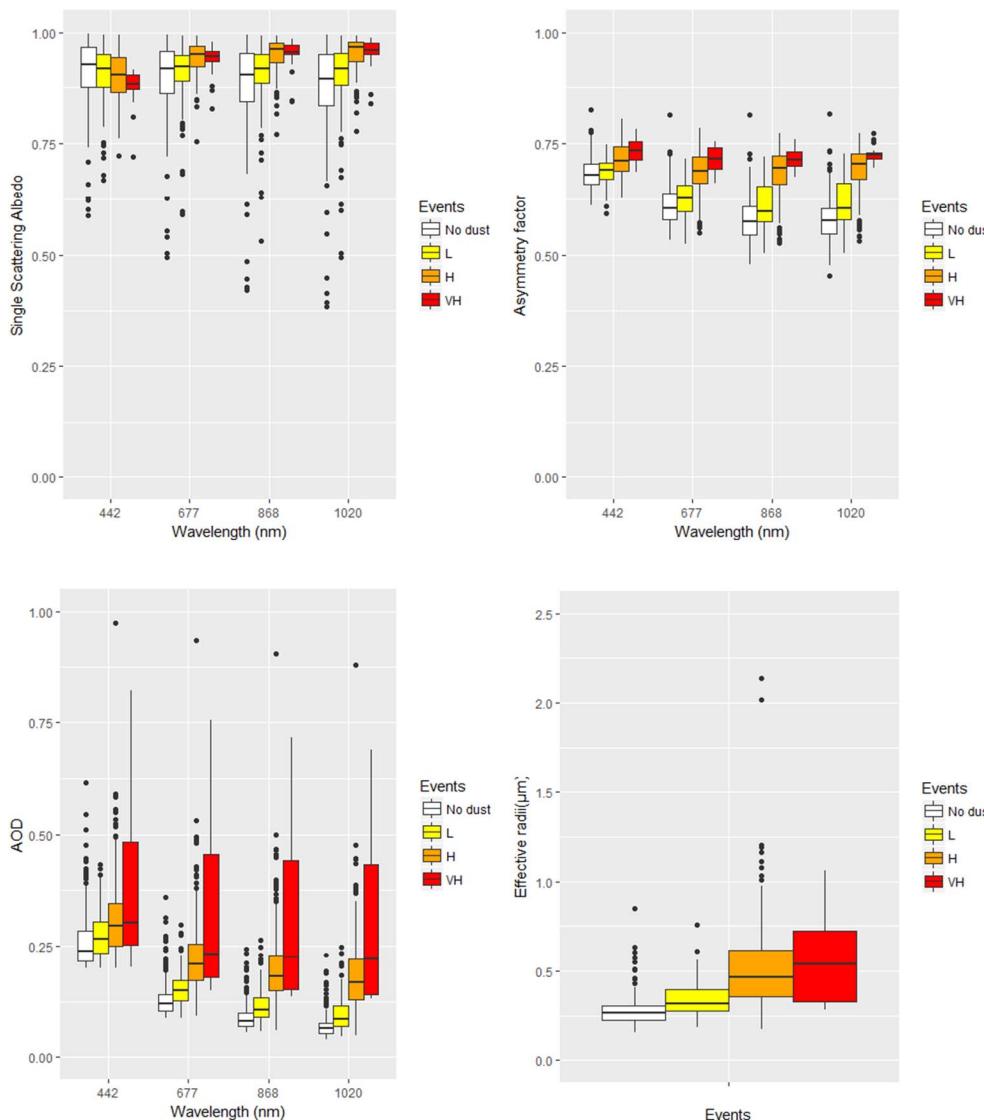


Fig. 3. Single scattering albedo and asymmetry factor at 442, 677, 868 and 1020 nm obtained in Madrid for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

enlarged (see Fig. 4 right). Finally, AOD seems to depend significantly on these events since the greater African dust contribution at ground level, the greater AOD. This trend is even clearer as the wavelength is enlarged. No Kolmogorov-Smirnov tests have been performed for SSA, asymmetry factor, AOD and effective radius because SSA depends basically on SAE and AAE and the tests were carried out for these two latter properties. Likewise, the results concerning AOD, effective radius and asymmetry factor follow the expected tendency produced by a contribution of coarse aerosols (except for SSA at 442 nm) and no further discussion was considered interesting.

Following on from this point, radiative forcing either at BOA and TOA (Fig. 5) seems to be more negative as the African dust contribution at ground level is greater. This again would be in agreement with the previous aerosol optical properties analyzed (SAE and AAE) where we concluded that African dust aerosols produce more scattering than typical Madrid aerosols which are more absorbent. The effect is observed clearer at BOA than at TOA. Statistically significant differences have also been found between “No dust”, “L”, “H” and “VH” in most of boxplots concerning the radiative forcing at BOA and TOA, except for those highlighted in red (Table 2), where the test indicates no statistical difference. The literature also suggests that the more intense the African dust event is the more negative the RF is. For instance, Barragán et al. (2017) found RF values of -42.3 W m^{-2} and -37 W m^{-2} at BOA in Granada and Lecce respectively under African dust conditions. In

addition, García et al. (2012b) also pointed out that mineral dust aerosols usually show more negative values of RF than aerosols classified as biomass burning and urban/industrial where black carbon is an important compound in these types of aerosol.

On another note, our analysis of RF^{eff} at BOA and TOA (Fig. 6) point out that as the African dust contribution at ground level is higher, the RF^{eff} is slightly higher (statistical differences are found in many cases, see Table 2) which is not expected. Given that intensive aerosol optical properties such as SAE, AAE and SSA have indicated before that African dust aerosols behave scattering more efficiently the radiation and they are also less absorbent in the shortwave, it is striking to find out higher RF^{eff} values during African dust events than under dust-free conditions as we expected them to be lower. On the other hand, if we considered that RF and RF^{eff} estimates are completely trustworthy we would have to conclude that the cooling effect at BOA and TOA caused by the African dust aerosol would be due to aerosol burden but not to their intensive aerosol optical property. Further research in this matter is necessary.

4.2. La Coruña

With regard to La Coruña site, SAE against AAE are represented in Fig. 7. It can be appreciated that these aerosol optical properties tend to be slightly more dispersed than those observed in Madrid under dust-

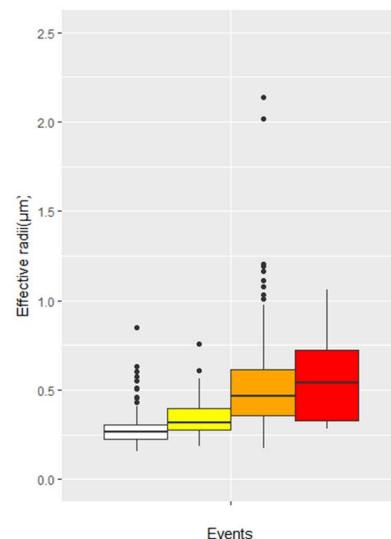


Fig 4. Aerosol optical depth (AOD) at 442, 677, 868 and 1020 nm and effective radii obtained in Madrid for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

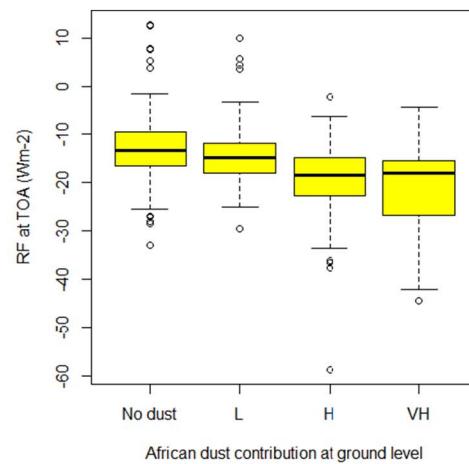
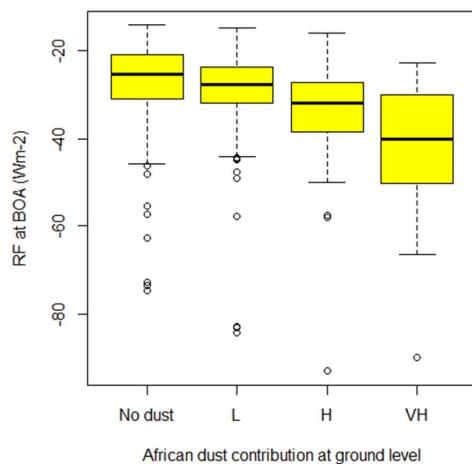
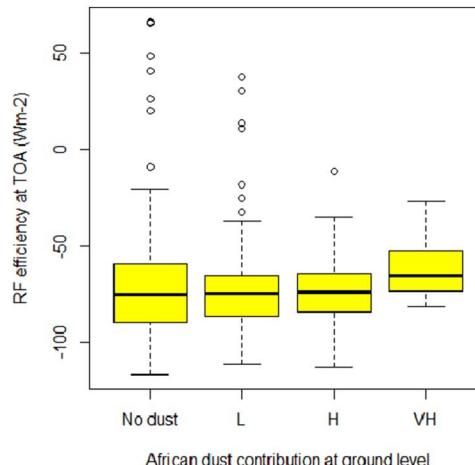
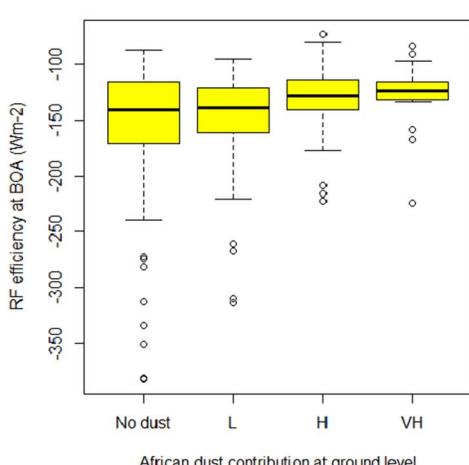


Fig. 5. Radiative forcing at Bottom Of Atmosphere (BOA) and Top Of Atmosphere (TOA) obtained in Madrid for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

free conditions. Mean values of SAE and AAE are 1.51 ± 0.34 and 1.09 ± 0.32 respectively. These values are also a little bit lower than SAE and AAE registered in Madrid under free-conditions. Moreover, it is noticeable that there is a minority group of data (in dust-free conditions) with really low AAE and SAE (Fig. 7) that are very different to the total population of data observed under dust-free conditions as most of them present values similar to the aforementioned mean values. We believe that the reason behind these exceptional cases is the fact that AOD is really close to 0.2. Other works have suggested earlier that under clean air conditions and low AOD, absorption process might be overestimated (Andrews et al., 2017). In Fig. 8, several cases which are representative of this situation are presented. So, the entrance of maritime clean air from the Atlantic would be responsible for low AOD and AAE. Likewise maritime aerosols are well known to have large size, which would result in a great capacity to scatter radiation and therefore a low SAE as well. As commented previously in the Experimental Sites section, the Comunidad Autónoma of Galicia, where La Coruña is located, is the region of Spain where more forest-fires occur, consequently these phenomena represent another important source of aerosol with distinct characteristics to the previous ones that might keep in mind. Finally, traffic is also significant aerosol sources at La Coruña. A possible explanation for the more dispersed value of SAE and AAE present in La Coruña than in Madrid for dust-free conditions can be due to a higher number of aerosol sources which in turn present different characteristics. In relation to SAE and AAE values, registered during African dust events in La Coruña, we have noticed that SAE (0.87 ± 0.53) tend to be a bit lower than SAE observed in Madrid under dusty conditions, however, the most remarkable observation is the fact that AAE obtained in La Coruña for dust conditions is



AAE vs SAE

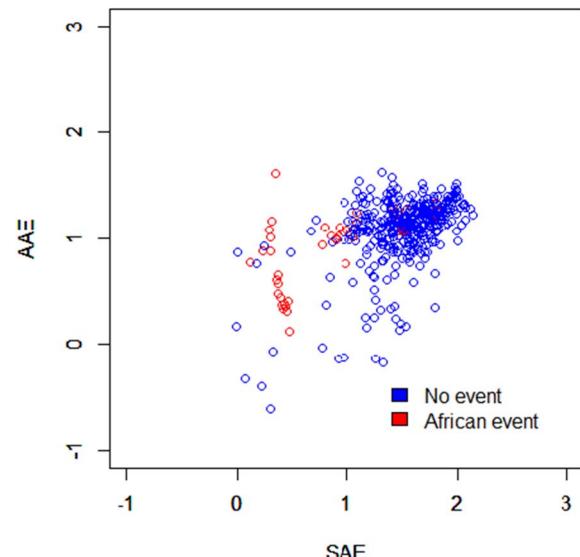


Fig. 7. Scattering and Absorption Angström exponent obtained in La Coruña for African desert dust (African event) and dust-free conditions (No event) studied from AERONET 1.5 level data.

0.91 ± 0.34 which is a really low value taking into account that these aerosols have their origin in the African dust transport. AAE values in Madrid and Granada respectively are 1.44 and 1.5 during these events.

Fig. 6. Radiative forcing efficiency at Bottom Of Atmosphere (BOA) and Top Of Atmosphere (TOA) obtained in Madrid for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

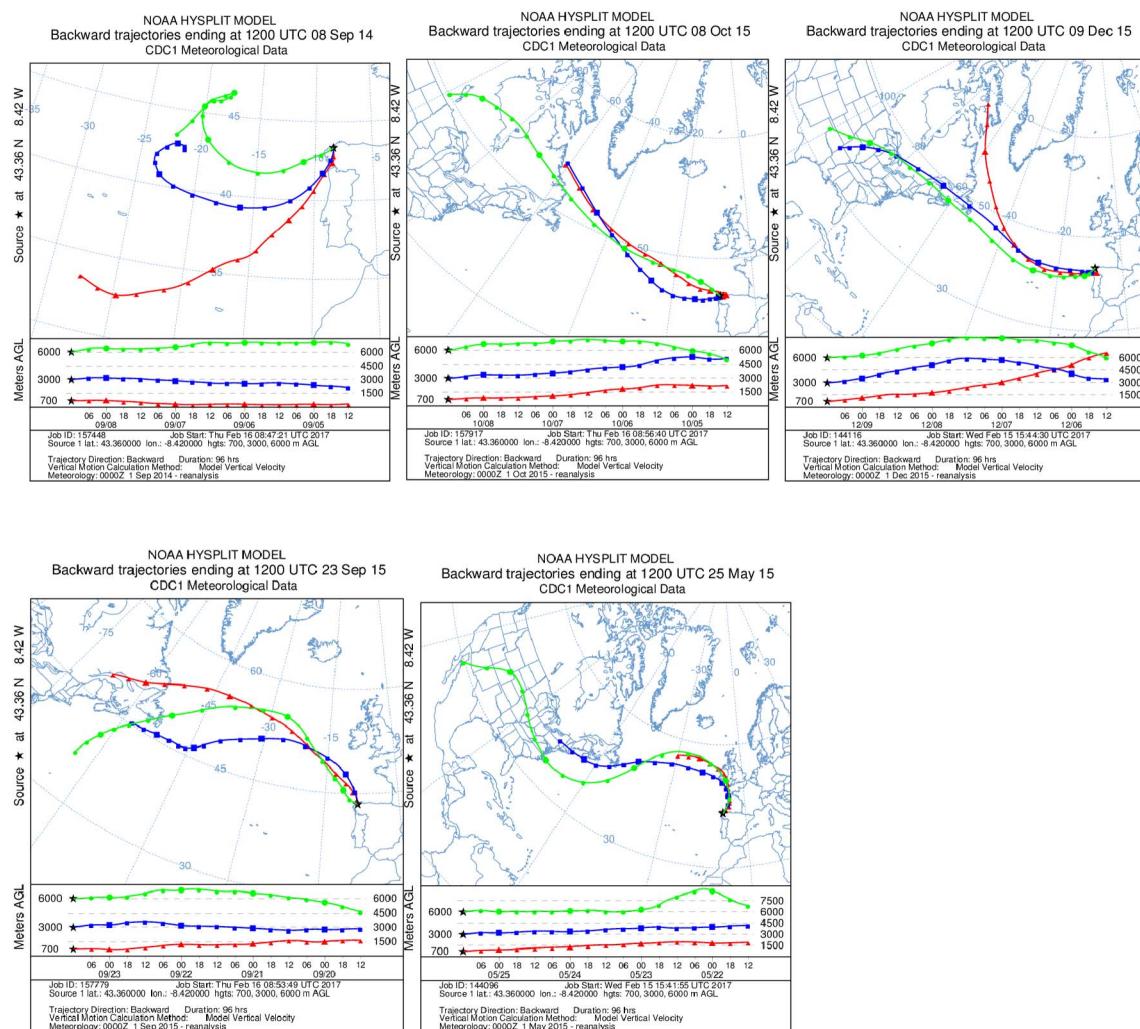


Fig. 8. Backward trajectories estimated by the HYSPLIT model at La Coruña site at 12 h UTC at altitude levels of 700, 3000 and 6000 m for the following days: 08/09/2014, 08/10/2015, 09/12/2015, 23/09/2015, 25/05/2015.

According to this property, these aerosols do have an absorbing-behavior at La Coruña which is not usual for typical African aerosol.

[Salvador et al. \(2014\)](#) demonstrated that African dust outbreaks over the western, central and eastern sides of the Iberian Peninsula are produced by different atmospheric circulation patterns. The main dust source areas are also different depending on the season of the year. In summer the prevailing transport of dust towards the central area of the Iberian Peninsula is produced from northern areas of Algeria, whereas it is produced from further regions, Western Sahara and southern Morocco, towards the western side of the Iberian Peninsula following an Atlantic pathway. Previous studies have also stated that African dust events over the Northwestern sector of the Iberian Peninsula are frequently produced in spring months due to the transport of dust plumes from the Sahel desert across the Atlantic Ocean following anticyclonic trajectories ([Querol et al., 2004](#); [Salvador et al., 2007](#)). Hence, the dust plumes have longer residence times over marine regions before reaching the Northwestern than the central sector of the Iberian Peninsula. In our study, 56% of African dust observations took place in spring time and 29% occurred in summer at La Coruña site ([Table 3](#)). By contrast, 70% of African dust observations were carried out in summer time whereas only 10% took place in spring at Madrid site ([Table 3](#)).

Hence it is suggested that African aerosol during transportation to La Coruña might have experienced an aging process under high humidity conditions, which should be responsible for their great capacity to absorb light. Nevertheless, the number of observations detected in La Coruña for dust conditions are relatively low as it can be noticed in

Table 3

Table 3
Distribution of African dust observations at each site from a seasonally standpoint: Spring (March, April, May), Summer (June, July, August), Autumn (September, October, November), Winter (December, January, February).

Site	Spring	Summer	Autumn	Winter
Madrid	10%	70%	17%	3%
La Coruña	56%	29%	15%	—

Fig. 7 and consequently it is difficult to state proper conclusions. Only 55 African dust measurements were taken in La Coruña against 354 African dust measurements performed in Madrid.

Concerning Fig. 9, we have observed that SAE and AAE values vary depending on the African dust contribution. The higher African dust contribution is, the low the SAE is (as expected) and the low AAE is. As stated before, it is striking to observe that as the African dust contribution at ground level is greater the AAE is lower. This means that this type of aerosols behaves absorbing light which is unexpected as African dust aerosols do not usually exhibit that pattern. We again state that this might be due to some aging/coating process suffered during the transport process.

SSA, asymmetry factor, and AOD at 442, 677, 868 and 1020 nm, and effective radius are presented in Figs. 10 and 11. No statistical tests have been performed for these aerosol optical properties due to the same reasons mentioned earlier for the Madrid station. When it comes

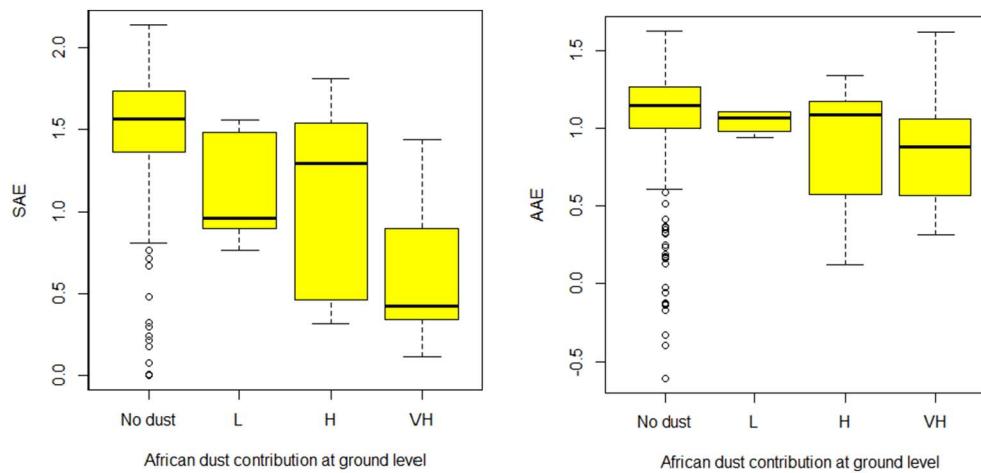


Fig. 9. Scattering and Absorption Angström exponent obtained in La Coruña for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

to studying the SSA, no trend has been found considering the African dust contribution at ground level. Unlike the SSA observed in Madrid, this is due to the fact that there is a counteraction between scatter and absorption processes. Both processes become greater as the African dust contribution at ground level is larger. Likewise, no qualitative difference in SSA is appreciated between dust-free and dust observations ("L", "H" and "VH"), perhaps, SSA at 868 and 1020 nm (dust-free conditions) might be slightly lower than SSA observed in dusty conditions. On another note, asymmetry factor at 677, 868 and 1020 nm is enlarged as the African dust contribution at ground level is greater. As observed previously in Madrid, this fact is attributed to aerosol size. In principle, African dust aerosols are usually comprised of large particles which should be responsible for a predominance of the scattering process in the forward direction. Therefore, if the contribution of African aerosol to the total mix of aerosols is greater it will be expected to observe a higher asymmetry factor. This trend is better observed as the wavelength is larger. Nevertheless, this fact has not been observed at 442 nm where asymmetry factor under dust-free conditions is nearly equal to the asymmetry factor in dust conditions (VH).

It is observed in Fig. 11 that AOD at 677, 868 and 1020 is higher as the African dust contribution at ground level is greater. This is due to the fact that African dust events tend to present a high aerosol burden which brings about a large AOD. Again, this observation is clearer as the wavelength is larger, although this is not observed for AOD at 442 nm. In a similar manner, effective radius is observed to increase as the African dust contribution at ground level is larger, however effective radius derived under dust-free conditions is greater than effective

radius obtained in African dust conditions (L) and nearly equal to African dust conditions (H). The comparison of the effective radius between Madrid and La Coruña (Figs. 4 and 11) indicates that effective radius under dust-free conditions seems to be similar at both places, however this is not observed under dusty conditions. The comparison of effective radius at (L, H and VH) dust conditions respectively at both places indicates that effective radius is slightly lower in La Coruña than in Madrid. It is suggested that this fact might be due to the distance from the source (African desert), in such a way that larger particles tend to be deposited earlier than smaller particles, so the longer distance transported by the particles, the smaller particles will be found in the total mix of aerosol (when considering African dust aerosols) as large particles are meant to be deposited throughout the way. Therefore, it has to be considered that Madrid is closer to the African mainland than La Coruña. As stated before, the source areas of dust and the transport pathways are different for the African dust outbreaks occurring over Madrid and La Coruña. Consequently, the characteristic of the mineral dust might differ between both regions, for example in terms of effective radius. Likewise, we would like to highlight that other differences have been also found for intensive optical properties such as SAE or AAE at both places (Figs. 2 and 9). For instance, SAE at African dust conditions (H and VH) are slightly lower in Madrid than in La Coruña or the AAE as a function of the African dust contribution at ground level behaves in an opposite manner at such places. Thus, the modification of African aerosol characteristics depends not only on a deposition process throughout the way but also on some aging or coating process as it was suggested before. So, when it comes to analyzing RF at BOA and TOA in

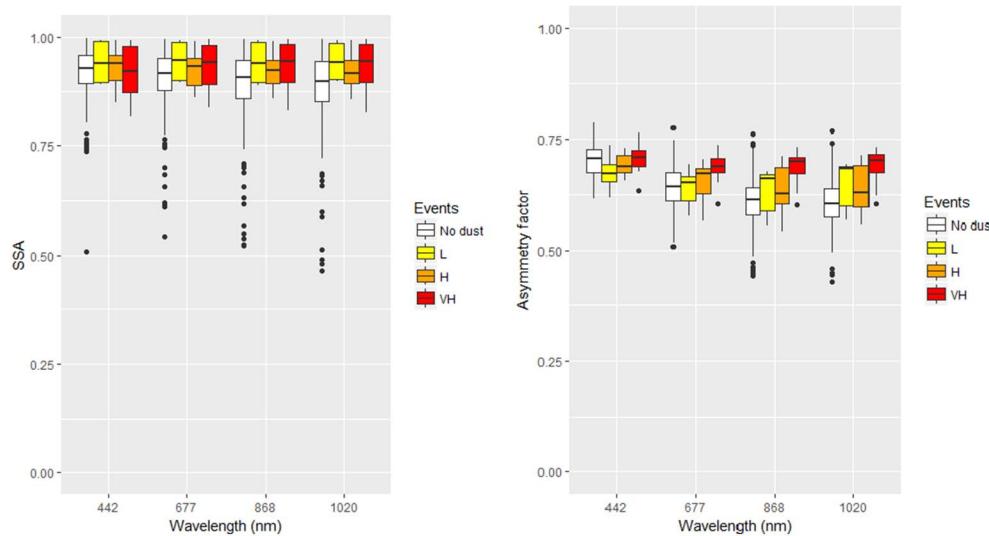


Fig. 10. Single scattering albedo and asymmetry factor at 442, 677, 868 and 1020 nm obtained in La Coruña for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

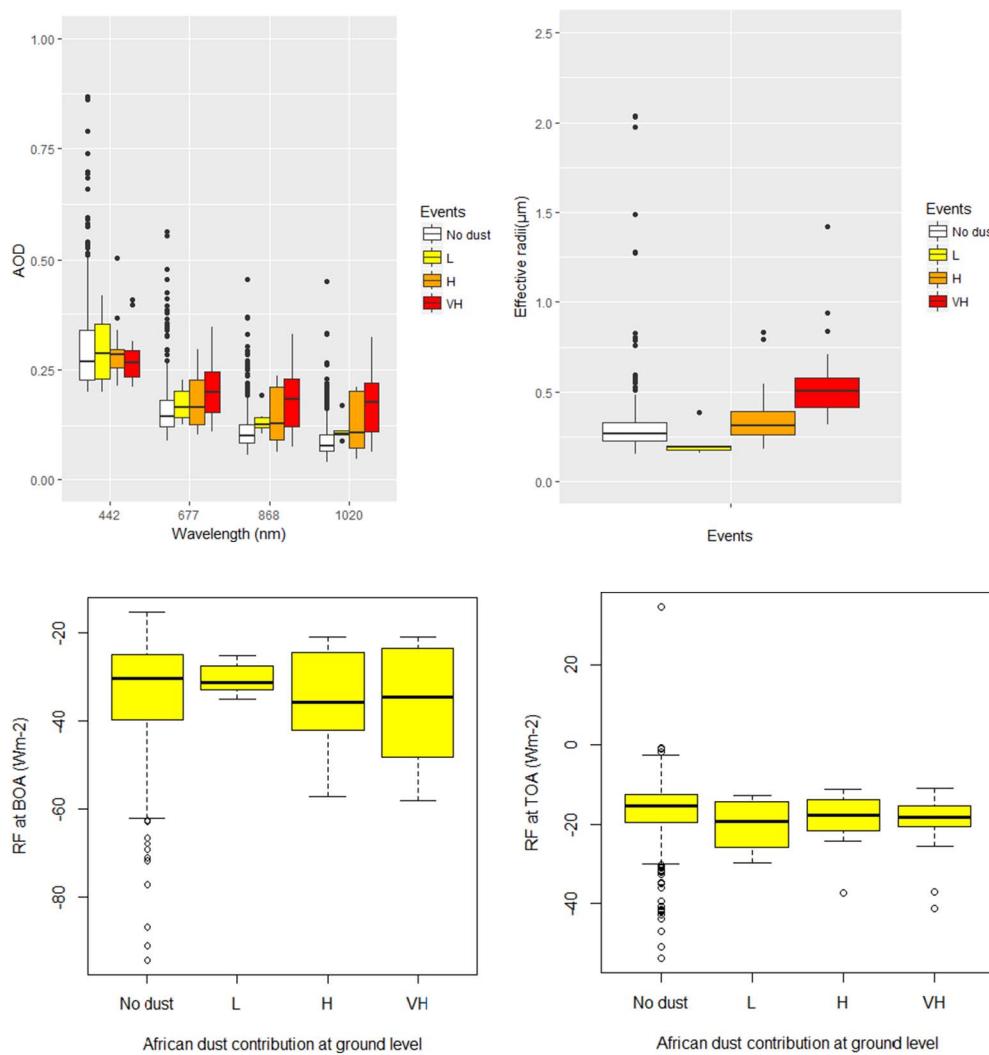


Fig. 11. Aerosol optical depth (AOD) at 442, 677, 868 and 1020 nm and effective radii obtained in La Coruña for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

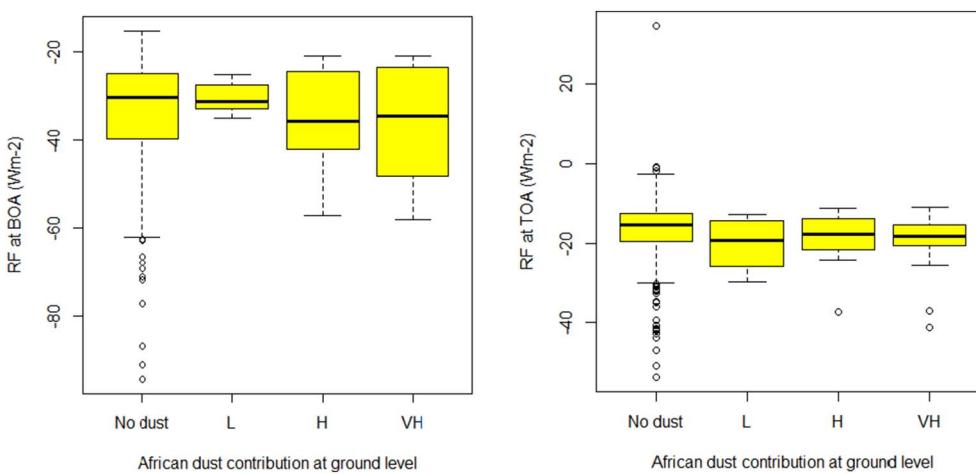


Fig. 12. Radiative forcing at Bottom Of Atmosphere (BOA) and Top Of Atmosphere (TOA) obtained in La Coruña for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

Table 4

The p values of the Kolmogorov-Smirnov statistical test applied to SSA, AAE, RF at BOA (efficiency), RF at BOA, RF at TOA (efficiency), RF at TOA for the La Coruña atmospheric column. The p values in red stand for those that are > 0.05 and consequently no statistical difference is found.

Aerosols property	T_1	T_2	T_3	T_4	T_5	T_6
SAE	1,05E – 02	2,98E – 03	0	6,65E – 01	2,62E – 02	1,38E – 03
AAE	2,35E – 02	2,81E – 01	1,06E – 04	3,02E – 01	8,20E – 02	2,99E – 02
RF at BOA (efficiency)	6,36E – 01	5,98E – 01	8,01E – 01	3,02E – 01	8,08E – 01	8,79E – 01
RF at BOA	3,41E – 01	3,78E – 01	3,29E – 01	1,82E – 01	1,38E – 01	4,16E – 05
RF at TOA (efficiency)	1,47E – 02	6,35E – 02	4,98E – 02	3,02E – 01	3,33E – 01	9,96E – 01
RF at TOA	3,08E – 01	3,10E – 01	1,85E – 02	4,60E – 01	4,70E – 01	7,07E – 01

La Coruña (Fig. 12), no trend is identified having considered the African dust contribution at ground level, neither is statistical difference between the distributions at the 95% confidence level in most cases (Table 4). Similarly, no trend and statistical difference has been found for the RF^{eff} at BOA and TOA in most cases (Fig. 13 and Table 4).

On another note, we performed a 2D cluster analysis based on SAE and AAE at both places to distinguish aerosols produced under distinct atmospheric conditions (dust-free and dusty conditions). The results of these analysis are shown in Fig. 14 only at Madrid, because poor results from a statistically point of view were found in La Coruña. Thus, it is clear that there are two groups of data in Fig. 14. Group 1 stands for those data that have been identified by the k-means method as AERONET observations carried out under dust-free conditions whereas group 2 relates to observations in African dust conditions. Group 1 presents mean values of AAE and SAE of 1.11 and 1.62 respectively

which are very similar to observational mean values in dust-free conditions (1.14 and 1.64 for AAE and SAE respectively). Group 2 reveals mean values of AAE and SAE of 1.74 and 0.48 which differ very little from observational mean values obtained during African events (1.44 and 0.96). The 2D cluster analyses has shown that it is an adequate technique to discern the two kind of aerosols in Madrid, given that mean values of African dust contribution at ground level is 56.63% ($\pm 21.63\%$) for group 2 (group meant to represent African dust aerosols) and 15.67% ($\pm 27.72\%$) for group 1 (group supposed to represent Madrid typical aerosols). What is more the ratio of between SS (Sum of Squares) to the total SS is 69.1%. This ratio is a measure of the goodness of the classification k-means. Therefore, we can state that the 2D cluster analysis performed in Madrid based on the SAE and AAE is valid. Besides, these properties have been proved to be useful in other places such as Granada (Valenzuela et al., 2015) which is even closer to the

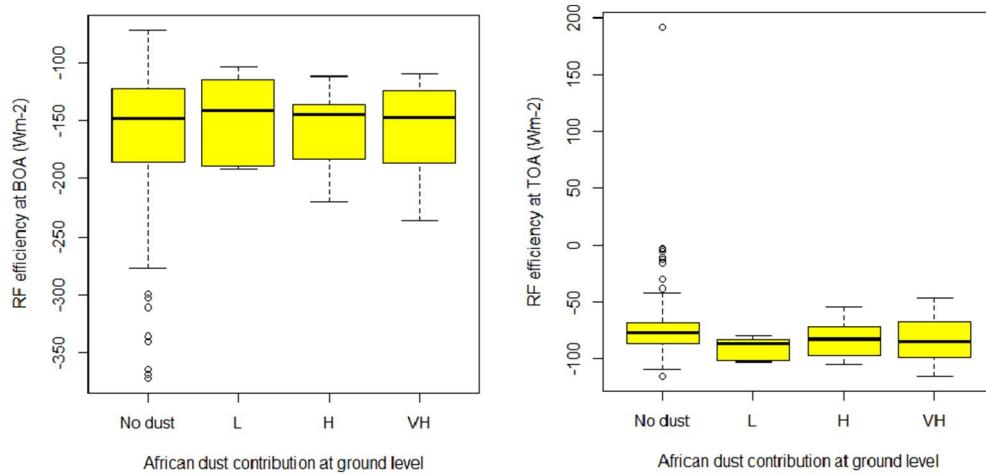


Fig. 13. Radiative forcing efficiency at Bottom Of Atmosphere (BOA) and Top Of Atmosphere (TOA) obtained in La Coruña for No dust, Low (L), High (H) and Very High (VH) African dust contribution at ground level studied from AERONET 1.5 level data.

African dust source. However we could not prove it for La Coruña site which is at a further distance than Granada and/or Madrid from the African dust aerosol source. We suggest again aerosol optical properties from African dust observed in La Coruña have changed due to the reasons commented earlier to that point that the 2D cluster analysis is not capable of establishing that distinction but it is for Madrid AERONET observations. Moreover, the observations carried out at La Coruña under African dust conditions are relatively low, and higher number of this kind observations should be necessary to come to a proper conclusion.

5. Conclusions

Aerosol measurements were studied at two AERONET sites of the Iberian Peninsula: Madrid and La Coruña so as to analyze aerosol optical properties along with RF, RF^{eff} estimates under dust-free and African dust conditions (L, H and VH). Mean values of SAE and AAE during dust-free conditions were found to be 1.64 ± 0.29 and 1.14 ± 0.23 respectively, which indicates a strong radiative-absorbent behavior (mainly due to black-carbon emissions) for this aerosol type in Madrid. On the contrary, mean values of SAE and AAE under African dust conditions were 0.96 ± 0.60 and 1.44 ± 0.51 and such mean values become lower than 0.5 and > 1.6 if African dust contribution at

ground level is larger than 50% of the PM_{10} total concentration. This fact indicates that aerosols observed during African dust conditions behave in an opposite manner from a radiative standpoint than Madrid typical aerosols as they produce greater radiation-scattering. In accordance with it, SSA at 677, 868 and 1020 nm tend to increase as the African event is more intense but not for SSA at 442 nm. Furthermore, effective radius, AOD and asymmetry factor reveal higher values as the African dust contribution at ground level is larger. The reason behind it is probably due to a high burden and size of aerosol during such events. In consequence, RF at BOA and TOA decrease as the African event is more intense which points out a cooling effect at Madrid site at this atmospheric layer. However RF^{eff} at BOA and TOA do not behave this way. Further research concerning RF^{eff} is necessary.

With regard to La Coruña site, the same methodology was applied to study the AERONET products, although there are less AERONET observations about dust events than in Madrid which hampers reaching solid conclusions, so a tentative explanation is given concerning La Coruña. The difference in aerosol optical properties and RF, RF^{eff} estimates under dust-free and African dust conditions were not so obvious for distinct reasons. We believe that under dust-free conditions, there are several aerosol sources though: maritime, traffic and combustion power plants and forest fires and consequently intensive aerosol optical properties do not show a specific pattern. What is more, SAE and AAE



Fig. 14. 2D cluster analysis based on Scattering Angström Exponent (SAE) and Absorption Angström Exponent (AAE). Event 1 and 2 stand for Madrid typical aerosol and African dust aerosol observed in the Madrid atmospheric column.

are reduced as the intensity of African event is larger. This latter property indicates that as the more intense the African event is, the greater absorption is produced which is not usual for such events at least at shortwave radiation. We believe that this fact might be due to some aging/coating process experienced by this aerosol kind. As a result no trend is identified in SSA as a function of the African dust contribution at ground level. Having considered the intensity of the African event Asymmetry factor, AOD and effective radius present similar pattern to the one observed in Madrid. However it is observed that effective radius at L, H and VH African dust contribution at La Coruña are slightly lower than in Madrid, which is attributed to a plausible deposition process since La Coruña is at a longer distance from the African dust source than Madrid. Given that aerosol properties of African dust might change dramatically by the time they reach La Coruña atmosphere no trend has been identified in RF and RF^{eff} estimates as function of the African dust contribution at ground level either at BOA and TOA. Finally, we suggest that when it comes to studying the RF and RF^{eff} produced by aerosols, the transport, the processes experienced by aerosols, and distance from the source is as important as the source itself on the aerosol kind. In this study we have pointed out that African dust aerosols might have different behavior from a radiative standpoint as they are at further distance from the source.

Acknowledgments

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