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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(λ)), 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°.45N, 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°.91N, 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.,