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Columnar and surface aerosol load over the Iberian Peninsula establishing annual cycles, trends, and relationships in five geographical sectors

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H I G H L I G H T S

• PM_{10} annual cycle shows two annual maxima; AOD only in the southern area.
• Air mass climatology explains the evolution of surface and columnar aerosols.
• Continuous fall in aerosol load is proved over the Iberian Peninsula (1985–2013).

A R T I C L E I N F O

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A B S T R A C T

The study of atmospheric aerosol load over the Iberian Peninsula (IP) under a climatological perspective is accomplished by means of PM_{10} and $AOD_{440\ nm}$ measurements from EMEP and AERONET networks, respectively, in the period 2000–2013. The PM_{10} annual cycles in five Iberian sectors show a main maximum in summer and a secondary maximum in spring, which is only observed in the southern area for the AOD climatology. The characteristics of PM_{10}–AOD annual cycles of each geographical sector are explained by the different climatology of the air mass origins and their apportioning. The two magnitudes are correlated with a factor ranging between 20 and 90 depending on the sector. The temporal evolution of the aerosol load has shown a notable decrease in the IP since the 1980s. Statistically significant trends are obtained in the Northeastern sector with a reduction of 26% (period 1985–2000) for the total suspended particles, which continues for the PM_{10} data with a value of 35% per decade (2001–2013), and also in the whole column, 61% per decade in the $AOD_{440\ nm}$ (2004–2013).
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1. Introduction

The effects caused by atmospheric aerosol particles on Earth's climate and radiative budget, air quality, human health and material degradation have motivated their analysis worldwide (e.g., Boucher et al., 2013, and the references therein). Climate change studies require a global and generalized aerosol characterization in order to assess its radiative forcing (direct and indirect effects; Lohmann and Feichter, 2005) and associated uncertainties. In order to successfully achieve this objective, aerosol measurements and modeling need to go together from global to regional scale. In the field of atmospheric aerosol studies, measurements based on remote sensing and "in situ" techniques have not always evolved together. Air quality and aerosol climate studies have constituted two traditional fields for aerosol studies with their specific particularities. The first one is usually linked to chemical–physical composition and pollution, while the second one is mostly related with the optical or radiative aerosol properties.

The primary element to account for the impact of the aerosol particles is their load in the atmosphere, represented by the mass concentration of particulate matter (PMx or TSP) in one case or by the aerosol optical depth (AOD) in the other one. The aerosol load in the atmosphere depends on both natural and anthropogenic emissions, atmospheric synoptic circulation patterns which govern long-range transport, local meteorology and topographical characteristics, among others. The PM_{10} (or $PM_{2.5}$) gives the particle mass concentration expressed in micrograms per unit of volume for particles with aerodynamic diameter less than 10 μm (or less than 2.5 μm). The TSPs refers to the total suspended particles. Although these variables can be measured at any atmospheric altitude by airborne platforms, the existing data series refer mainly to ground-based records belonging to different air quality

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networks. Besides, the aerosol optical depth (AOD) represents a dimensionless parameter linked to the aerosol load in the whole atmospheric column, as an indicator of the radiation attenuation by aerosol particles.

The optical or radiative aerosol properties are measured by global world wide networks, as the AERONET (Aerosol Robotic Network, http://aeronet.gsfc.nasa.gov), or the GAW-PFR network (http://www.pmodwrc.ch/worcc). These networks are based on sun-photometer instruments which measure direct solar and diffuse sky radiation at several wavelengths, being the spectral AOD the primary key property. The AERONET project, starting in the 1990s, has been developed with the aim of monitoring the long term aerosol optical properties from ground, as well as providing a global database for the establishment of aerosol climatology, validation of satellite retrievals and aerosol models. Several local aerosol climatologies have been established based on this type of data (see, e.g., Holben et al., 2001; Dubovik et al., 2002; Toledano et al., 2007; Bovchaliuk et al., 2013). AERONET is structured as a federation of regional–national networks, among them the Iberian Network for Aerosol Measurements (RIMA, http://www.rima.uva.es), created in 2004 and managed by the Group of Atmospheric Optics, University of Valladolid (GOA-UVA). The RIMA–AERONET network ensures the provision of long-term and quality controlled data at several locations across Spain, Portugal and other countries in Europe into the AERONET-Europe framework.

Aerosol measurements in remote (background) sites are a useful tool to establish the aerosol long-term trends. For this purpose, the European Monitoring and Evaluation Programme (EMEP) was created as a scientifically based and policy driven program under the Convention on Long-range Transboundary Air Pollution (CLRTAP) for international co-operation. In the EMEP report of 2013 (Aas et al., 2013), the yearly evolution of PM_{10} emissions was established between 2000 and 2011 for the European continent. There are countries with a notable increase of PM_{10} emissions during this period (e.g., Belarus, Moldova, Bulgaria, and Lithuania). However, other European regions present a decrease of PM_{10} emissions (e.g., Belgium, France, The Netherlands, and UK). In particular, Spain has shown a decrease around 25%. Furthermore, the PM_{10} mass measurements from sixteen European sites have shown an average decrease of 18% per year between 2000 and 2011, which means an annual loss (in average mass concentration) of 0.29 $\mu g\ m^{-3}$ (Aas et al., 2013). The directive 2008/50/EC of the European Parliament (May 2008) aims at reducing pollution and improving the air quality over Europe limiting the thresholds that can be reached at different time scales. For instance, to protect human health, the daily mean PM_{10} concentration must not exceed 50 $\mu g\ m^{-3}$ more than 35 times through a year, being the annual mean limited to 40 $\mu g\ m^{-3}$.

The interest of the Mediterranean Basin and, particularly, Spanish region is motivated by the high PMx concentration compared to other European regions. The Mediterranean Basin is a vulnerable region in terms of the climate destabilization, which produces a more complex role of the aerosol particles in the climate system (Querol et al., 2009a). Many studies have focused on the particulate matter evolution in Spain or Western Mediterranean area (e.g., Querol et al., 2009a, 2009b; Barmpadimos et al., 2012; Salvador et al., 2012; Cusack et al., 2012; Pey et al., 2013; Gkikas et al., 2013; Bennouna et al., 2014; Notario et al., 2014). For instance, Salvador et al. (2012) have shown that the PM anthropogenic sources have decreased between 1998 and 2008 in the Madrid metropolitan area (Central Spain). For the same latitudinal belt, Notario et al. (2014) reported the weekend effect for PM_{10} data. Overall, PM concentration seems to decrease in the last years in the Iberian Peninsula (e.g., Cusack et al., 2012; Barmpadimos et al., 2012). The seasonal cycle followed by PM concentrations with large values during the warm season is shown by, e.g., Querol et al. (2009a), Kassomenos et al. (2014), and Notario et al. (2014).

Several studies have dealt with the high influence played by African mineral dust events in the Mediterranean Basin (e.g., Querol et al., 2009a, 2009b; Pey et al., 2013; Gkikas et al., 2013). Overall, 15% of the PM_{10} levels over Spain is due to desert dust events (Querol et al., 2009b). Concerning human health, Reyes et al. (2014) have shown that periods under African air mass intrusions presented a significant increase in respiratory-cause admissions in hospitals of Madrid (Central Spain).

The geographical position of the Iberian Peninsula makes that this area is reached by a variety of air masses of various origins (maritime, continental and tropical, among others), with a prevalence of air masses of maritime (Atlantic) character (e.g., Valverde et al., 2014). Toledano et al. (2009) obtained, from an air mass analysis over South-western Spain, that Saharan dust intrusions are originated from North Africa as tropical continental and maritime air masses mostly in summer and late-winter/early spring. For the central area, Salvador et al. (2013) found a larger contribution of the Atlantic origin, while the Mediterranean and Continental (European) source areas seem to play a minor role. For central Spain, desert dust intrusions occur on 9% of the days between 2001 and 2008.

With this background, the aim of this study is to perform a meaningful analysis of the aerosol load over the Iberian Peninsula (IP), considering both PMx and AOD data from a climatological perspective. We are aware that the use of the term "climatology" for the aerosol data series in the IP must be taken with care. However, the existing series give a valuable view about the behavior of the annual cycles and inter-annual changes. Derived parameters as the $PM_{2.5}/PM_{10}$ ratio and the Ångström α coefficient, which provide information on the fine or coarse particle predominance, are also considered in the analysis. Particulate matter concentration (PM_{10} and $PM_{2.5}$) at the surface is obtained from the EMEP Spanish stations, while the aerosol optical depth data are taken from all the available AERONET (AErosol RObotic NETwork) Iberian sites. The analyzed period spans from 2000 to 2013 (data availability depends on the sites). The simultaneous use of the surface and columnar aerosol measurements is a suitable approach to characterize the aerosol load, but for this purpose, a detailed air mass analysis is also required. In this context, five Iberian sectors have been considered because of the observed differences among all the sites. The contribution of seven types of air masses has been evaluated. The relationship between the surface and columnar aerosol is also investigated. Finally, the study is complemented with the long-term trends of the aerosol load in the last quarter century. The total suspended particles (TSPs) in the period 1988–2000 were included to extend the analysis of surface aerosols.

This study is affordable in the Iberian Peninsula due to the large density in aerosol ground-based sites which is not easily encountered in other worldwide countries/areas. The use of geographical sectors minimizes the existence of temporal gaps in the individual data series caused by different problems. The analysis of the aerosol properties from a global perspective helps in understanding the large variety and variability of aerosol conditions in the small latitudinal belt of 8° spanned by the Iberian Peninsula. Furthermore, the knowledge of the long-term trends and climatology of the aerosol load is required to implement new strategies in air quality control. In this sense, the simultaneous analysis of PMx and AOD provides a more complete characterization of the aerosol particles in the whole atmospheric column.

2. Database and methods

2.1. Data collection

The aerosol optical depth (AOD) data used in this study are collected from AERONET (AErosol RObotic NETwork) (Holben et al., 1998) in all the Iberian sites with data during, at least, one year (see Table 1 and Fig. 1a). In this study, the AOD at 440 nm wavelength ($AOD_{440\ nm}$) and Ångström exponent (α) are level 2.0 (quality assured) data. The radiometers placed at the Iberian Peninsula are part of RIMA and are

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