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Aerosol properties over the western Mediterranean Basin: temporal and spatial variability

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Abstract

This study focuses on the analysis of AERONET aerosol data obtained over Alborán Island (35.95° N, 3.01° W, 15 m a.s.l.) in the western Mediterranean from July 2011 to January 2012. Additional aerosol data from three nearest AERONET stations and the Maritime Aerosol Network (MAN) were also analyzed in order to investigate the aerosol temporal and spatial variations over this scarcely explored region. Aerosol load over Alborán was significantly larger than that reported for open oceanic areas not affected by long-range transport. High aerosol loads over Alborán were mainly associated with desert dust transport from North Africa and occasional advection of anthropogenic fine particles from Italy. The fine particle load observed over Alborán was surprisingly similar to that obtained over the other three nearest AERONET stations in spite of the large differences in local aerosol sources. The results from MAN acquired over the Mediterranean Sea, Black Sea and Atlantic Ocean from July to November 2011 revealed a pronounced predominance of fine particles during the cruise period. Alborán was significantly less influenced by anthropogenic particles than the Black Sea and central and eastern Mediterranean regions during the cruise period. Finally, the longer AERONET dataset from Málaga (36.71° N, 4.4° W, 40 m a.s.l.), port city in southern Spain, shows that no significant changes in columnar aerosol loads since the European Directive on ship emissions was implemented in 2010 were observed over this site.

1 Introduction

Atmospheric aerosol particles play an important role in the atmosphere because they can affect the Earth's radiation budget directly by the scattering and absorption of solar and terrestrial radiation (e.g., Haywood and Shine, 1997), and indirectly by modifying cloud properties (e.g., Kaufman et al., 2005), and hence have important climate implications. Understanding the influence of atmospheric aerosols on radiative transfer in the atmosphere requires accurate knowledge of their columnar properties, such as

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the spectral aerosol optical depth a property related to aerosol amount in atmospheric column (Haywood and Boucher, 2000; Dubovik et al., 2002). Global measurements of columnar aerosol properties including spectral aerosol optical depth can be assessed from satellite platforms (e.g., Kaufman et al., 1997). However, satellite aerosol retrievals suffer from large errors due to uncertainties in surface reflectivity, and furthermore have low temporal resolution. Currently, the ground sun photometric technique is considered the most accurate for the retrieval of aerosol properties in the atmospheric column. Thus, many ground based observation networks have been established in order to understand the optical and radiative properties of aerosols and indirectly evaluate their effect on the radiation budget and climate (e.g., AERONET). However, quantification of aerosol effects is very difficult because of the high spatial and temporal variability of aerosol physical and optical properties (Forster et al., 2007). This high aerosol variability is due to their short atmospheric lifetime, aerosol transformations, aerosol dynamics, different meteorological characteristics, and the wide variety of aerosol sources (Haywood and Boucher, 2000; Dubovik et al., 2002). In this sense, Forster et al. (2007) highlighted the large uncertainties on the aerosol impact on radiation budget. Therefore, monitoring of aerosol properties at different areas in the world can contribute to reduce these uncertainties.

Most of the planet is covered by Oceans and Seas, and thus the study of marine aerosol is a topic of ongoing interest (e.g., Smirnov et al., 2002). Particularly, many efforts are being made to characterize this aerosol type from ground based measurements, leading to the creation of the Maritime Aerosol Network (MAN) as part of the AERONET network (Smirnov et al., 2009). However, MAN lacks continuous temporal measurements, and thus measurements from remote islands in the Oceans and Seas are required. Particularly, in the Mediterranean Basin aerosol properties are characterized by a great complexity, due to the presence of different types of aerosols such as maritime aerosols from the Mediterranean Sea itself, biomass burning aerosols from forest fire, anthropogenic aerosols transported from European urban and industrial areas, mineral dust originated from north African arid areas, and anthropogenic particles emitted from the intense ship traffic in the Mediterranean Sea (e.g., Lelieveld et al., 2002; Barnaba and Gobbi, 2004; Lyamani et al., 2005; Lyamani et al., 2006a, b; Papadimas et al., 2008; Viana et al., 2009; Pandolfi et al., 2011; Alados-Arboledas et al., 2011; Becagli et al., 2012; Valenzuela et al., 2012a, b). Past studies revealed that the aerosol load and the aerosol direct radiative effect over the Mediterranean are among the highest in the world, especially in summer (e.g., Leleived et al., 2002; Markowicz et al., 2002; Papadimas et al., 2012; Antón et al., 2012).

In this framework, the characterization of aerosol over the Mediterranean has received great scientific interest. To date, a large number of studies has been done focusing on the eastern and central regions (e.g., Formenti et al., 1998; Balis et al., 2003; Gerasopoulos et al., 2003; Di Iorio et al., 2003; Kubilay et al., 2003; Pace et al., 2005, 2006; Fotiadi et al., 2006; Meloni et al., 2007, 2008; Di Sarra et al., 2008; Di Biagio et al., 2010; Boselli et al., 2012). However, few studies have been done in the western Mediterranean Basin (Horvath et al., 2002; Alados-Arboledas et al., 2003; Mallet et al., 2003; Estelles et al., 2007; Saha et al., 2008; Pérez-Ramírez et al., 2012, Foyo-Moreno et al., 2014). The majority of these studies have been performed in coastal Mediterranean urban sites largely influenced by local pollution emissions, except those carried out at Crete and Lampedusa islands in the eastern and central Mediterranean Sea regions. In general, columnar aerosol data are scarce over the Mediterranean Sea and almost absent over the western Mediterranean Sea. Thus, measurements of the aerosol properties over the western Mediterranean Sea are needed in order to evaluate the aerosol regimes over this scarcely explored region (Smirnov et al., 2009). In order to fill this gap and provide columnar aerosol properties over the western Mediterranean Sea, the Atmospheric Physic Group of the University of Granada, Spain, in collaboration with Royal Institute and Observatory of the Spanish Navy (ROA) has installed a sun photometer at Alborán; a very small island in the western part of Mediterranean Sea located midway between the African and European continents. Currently, this station is part of AERONET network (http://aeronet.gsfc.nasa.gov).

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Ship emissions have become increasingly important sources of air pollution in the Mediterranean Basin, and are expected to increase and equal or even exceed those from land sources in the future (Cofala et al., 2007). Ships are particularly important sources of SO_2 and indirectly also of sulphate aerosol, formed by oxidation of SO_2 in the atmosphere. Indeed, Marmer and Langmann (2005) have estimated that 54 % of the sulphate aerosol load in the atmospheric column over the Mediterranean Sea originates from ship emissions, contributing over 50 % to the direct radiative forcing in the Mediterranean. In addition, these authors showed that the sulfate aerosol load in the atmospheric column over the Mediterranean Sea is remarkably higher than over European land in summer. Furthermore, Marmer and Langmann (2005) suggest that the reduction of sulphur content in ship fuel would substantially reduce the total aerosol loads and thus aerosol radiative forcing in the Mediterranean.

As a result of the growing role of ship emissions in the Mediterranean, the European Union has implemented some regulations in order to minimize its potential environmental and climatic effects. In this sense, the European Union directive (Directive 2005/33/EC) requires that all ships at berth or anchorage in European harbors use fuels with a sulphur content of less than 0.1 % by weight since 1 January 2010. Prior to this date, ships in the Mediterranean Basin were allowed to use fuels with up to 4.5 % sulphur content. Thus, the implementation of this directive is expected to have a significant impact on columnar aerosol loads over the Mediterranean Basin (Marmer and Langmann, 2005). This can have large impacts on the atmospheric aerosol properties normally observed over the Mediterranean Sea and can lead to a large change of radiative fluxes at surface, atmosphere and top of atmosphere. To our knowledge, there are no studies in the literature about the impact of this new European Directive on the columnar atmospheric aerosol properties over Mediterranean Basin. Thus, long term sun photometer measurements from Mediterranean port cities or Island sites provide an excellent opportunity for examining the impact of the European Directive on aerosol characteristics, since they are expected to be strongly influenced by emissions from ships.

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Measurements of aerosol properties are lacking in the western Mediterranean marine region. This study focuses on the characterization of aerosol load and aerosol types (sizes) as well as their temporal variability over Alborán Island in the western Mediterranean from July 2011 to January 2012. In addition, special attention is given to investigate the conditions responsible for large aerosol loads over this Island and much attention is paid to identify the potential aerosol sources affecting Alborán. Furthermore, additional aerosol properties from three AERONET stations surrounding Alborán Island and from MAN cruise are analyzed here to investigate the spatial aerosol variation over the Mediterranean Basin. Finally, we investigate the possible impact of the implantation of European ship emission regulations on the columnar aerosol load and types over western Mediterranean. To this end, a longer time series of AERONET data from Málaga port city, which is expected to be influenced by ship emissions, is investigated here.

The work is structured as follows. In Sect. 2 we describe the instrumentation used and the experimental sites. Section 3 is devoted to the main results, where we analyze the aerosol optical properties at Alborán Island, the spatial variability of aerosol properties in the Mediterranean and the impact of the European ship emission regulations on columnar aerosol properties over Málaga AERONET station. Finally, in Sect. 4 we present the summary and conclusions.

2 Instrumentation and study sites

This study focuses on the AERONET sun photometer measurements acquired at the Alborán Island (35.95° N, 3.01° W, 15 m a.s.l.), in the western Mediterranean Sea, from July 2011 to January 2012. Alborán is a small island with an approximate surface of 7 ha, located ∼ 50 km north of the Moroccan coast and 90 km south of the Spanish coast (Fig. 1). Currently, only 12 members of a small Spanish Army garrison live on the island. The island and its surrounding area are declared a natural park and marine reserve. There are no local anthropogenic emission sources at Alborán; however, the island is just south of an important shipping route. Due to its location, Alborán Island is expected to be affected, depending on the regional circulation, by anthropogenic pollutants originated in urban and industrial European areas and also by desert dust transported from North African arid regions. The climate of the region depends strongly on the Azores anticyclone. Winter is mainly characterized by low pressure passing over the Iberian Peninsula, resulting in the prevalence of westerly winds and enhanced rainfall. In this season, the weather is unstable, wet and windy. In summer, the well-established Azores high produces dry and mild weather with easterly winds that combine with sea/land breezes created by the aridity of the coastal mountains (Sumner et al., 2001).

Columnar aerosol properties were measured by a CIMEL CE-318-4 sun photometer, which is the standard sun photometer used in the AERONET network (Holben et al., 1998). This instrument has a full view angle of 1.2° and makes direct sun measurements at 340, 380, 440, 500, 670, 870, 940 and 1020 nm (nominal wavelengths). The direct sun measurements are then used to retrieve the aerosol optical depth at each wavelength, $\delta_a(\lambda)$, except for 940 nm which is used to compute precipitable water vapor (Holben et al., 1998). Detailed information about the CIMEL sun photometer can be found in Holben et al. (1998). The total estimated uncertainty in $\delta_a(\lambda)$ provided by AERONET is of ±0.01 for $\lambda > 440$ nm and ±0.02 for shorter wavelengths (Holben et al., 1998). Furthermore the spectral dependency of the $\delta_a(\lambda)$ has been considered through the Angström exponent, α(440–870), calculated in the range 440–870 nm. The Angström exponent provides an indication of the particle size (e.g., Dubovik et al., 2002). Small values of the Angström coefficient ($\alpha(440\text{–}870) < 0.5$) suggest the dominance of coarse particles, such as sea salt or dust, while $\alpha(440\text{–}870) > 1.5$ indicates the predominance of small particles such as sulphate, nitrate and biomass burning particles. Also included in the analysis are the fine mode (δ_F(500 nm)) and coarse mode (δ_C(500 nm)) aerosol optical depths at 500 nm as well as the fine mode fraction (FMF) (ratio of δ_F(500 nm) to δ_a(500 nm)), determined using the spectral de-convolution algorithm method developed by O'Neill et al. (2003).

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In addition, to investigate the spatial variation of aerosol properties over the western Mediterranean, we used AERONET data from three AERONET stations surrounding Alborán Island; Oujda, Málaga and Palma de Mallorca (see Fig. 1). These sites cover different environments including, urban, coastal and island sites and have different background aerosol characteristics. Palma de Mallorca (39.35° N, 2.39° E, 13 m a.s.l.), the capital of the Balearic Islands, is the largest city in the Mallorca Island with a population of around 400 000. It is located in the western Mediterranean Sea, about 250 km from the African continent and 190 km from the Spanish coast. Málaga (36.72° N, 4.5° W, 40 m a.s.l.), with a population of around 600 000 is the major coastal city in southeast Spain on the Mediterranean coast. Oujda city (34.65° N, 1.89° W, 450 m a.g.l.) is located in eastern Morocco, 60 km south of the Mediterranean Sea, with an estimated population of 450 000. In this study, the level 2 AERONET aerosol data are used.

Furthermore, we used ship sun photometer measurements collected on-board the *Nautilus_11* on the Mediterranean Sea, Atlantic Ocean and Black Sea during the period July–November 2011. These measurements were made in the framework of the Maritime Aerosol Network (MAN), a component of AERONET (Smirnov et al., 2011). Information about the Nautilius_11 cruise track can be found in (http://aeronet.gsfc.nasa.gov/new_web/cruises_new/Nautilus_11.html). MAN uses Microtops II sun photometers and utilizes calibrations and data processing procedures of AERONET network. The Microtops II sun photometer used in this cruise acquires direct sun measurements at 440, 500, 675 and 870 nm. The estimated uncertainty of the optical depth in each channel is around ±0.02 (Knobelspiese et al., 2004). Level 2 MAN data are used in this study.

To characterize the transport pathways and the origins of air masses arriving at our studied sites, 5 day backward trajectories ending at 12:00 UTC at these sites for 500, 1500, 2500, 3500, 4500 and 5000 m above ground level were calculated using the HYSPLIT model (Draxler and Rolph, 2003). The model version employed uses CDC1 Meteorological data and includes vertical wind.

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3 Results and discussion

3.1 Temporal evolution of aerosol properties over Alborán Island

Figure 2 shows the temporal evolutions of daily mean values of aerosol optical depths at at 340, 500, and 1020 nm and α(440–870) measured at Alborán Island in the western Mediterranean from July 2011 to January 2012. There are some gaps in $\delta_a(\lambda)$ and α(440–870) data series due to some technical problems and the presence of clouds (invalid data). Table 1 presents a statistical summary of daily average values of all the analysed aerosol properties. One of the main features observed is the large variability of $\delta_a(\lambda)$ (for example, δ_a(500 nm) ranged from 0.03 to 0.54) that is primarily related to changes in the air masses affecting the study area, as can be seen hereafter. The coefficient of variation (COV), defined as the standard deviation divided by the mean value, can be used to compare the variability of different data sets. As shown in Table 1, the $\delta_a(\lambda)$ at 1020 nm (with COV of 91 %) showed much greater variability than at 340 nm (with COV of 60 %). It is well known that $\delta_a(\lambda)$ at higher wavelengths is more affected by naturally produced coarse particles (radius above 0.5 µm) like dust and sea salt particles, while $\delta_a(\lambda)$ at smaller wavelengths is more sensitive to the fine particles (radius below 0.5 µm) such as those from anthropogenic activities or biomass-burning. Thus, the higher variability of $\delta_a(\lambda)$ for large wavelengths indicates strong variability in the coarse particle load (dust or sea salt) over Alborán Island. This result is also supported by the larger COV of δ_C(500 nm) as compared to δ_F(500 nm) (Table 1). Aerosol salt emission variations due to the wind speed variation and the changes in the frequency and intensity of dust intrusions over the Island may explain the large variability in the coarse particle component and hence the large $\delta_a(\lambda)$ variability for large wavelengths. Moreover, coarse particles have shorter residence time in the atmosphere in comparison with fine particles, which could explain also the large $\delta_C(\lambda)$ variability. On the other hand, α(440–870) values also show large variability and varied from 0.2 to 1.7 with mean value of 0.8 ± 0.5, indicating different atmospheric conditions dominated by different aerosol types (coarse particles, fine aerosols and different mixtures of both coarse and fine particles). It is noted that on 70 % of the analysed days the values of α(400–870) were lower than 1, suggesting that coarse particles dominated the aerosol population over the Alborán Island for most of the analysed days. This is further supported by the analysis of fine mode fraction which ranged from 0.20 to 0.90 (mean value of 0.47 ± 0.15); with daily mean values less than 0.5 on 65 % of the analysed days.

The observed mean δ_a(500 nm) value over Alborán Island was significantly higher (by factor of 2) than that reported by Smirnov et al. (2002) (δ_a(500 nm) in the range 0.06–0.08) for open oceanic areas not affected by long-range aerosol transport. Moreover, the mean δ_a(500 nm) and α(440–870) values obtained in this study were larger than the global mean δ_a(500 nm) value of 0.11 and α(440–870) of 0.6 reported for maritime aerosols by Smirnov et al. (2009). On other hand, average aerosol optical depths at 495.7 nm of 0.24 ± 0.14 and α(415–868) of 0.86 ± 0.63 were obtained from multi filter rotating shadowband radiometer at Lampedusa Island (in the central Mediterranean Sea) during July 2001–September 2003 (Pace et al., 2006). Using AERONET data measured in Crete Island (eastern Mediterranean Sea) during 2003–2004, Fotiadi et al. (2006) have reported mean δ_a(500 nm) value of 0.21 and α(440–870) of 1.1. The differences between aerosol properties observed over Alborán, Lampedusa and Crete Islands could be due, at least partly, to the different measurement periods and the methodologies employed. Later we compare the results obtained over Alborán to those observed (during the same period and using the same type of instruments) over three nearby AERONET stations.

According to the Smirnov et al. (2002) criterion, background maritime conditions can be generally found when δ_a(500 nm) < 0.15 and α(440–870) is lower than 1. Considering this criterion, background maritime conditions were observed over Alborán Island only on 40 % of the analyzed days. According to back trajectory analysis, almost all these days were characterized by advection of clean Atlantic air masses over the study area. In addition, the majority of these background maritime cases were observed from November to January. This result is in agreement with the study performed at the Island of Lampedusa, in the central Mediterranean, showing that background maritime conditions are usually observed during Atlantic air advection (Pace et al., 2006). However, clean Maritime conditions observed over Alborán Island during the analyzed period are more frequent than those observed over the central Mediterranean Sea. Pace et al. (2006) have shown that clean maritime conditions are rather rare over the central Mediterranean due to the large impact of natural and anthropogenic sources. The difference in the frequencies of clean Maritime conditions at these two sites can be explained by their different locations. Alborán is closer to the Atlantic Ocean than is Lampedusa, and the Atlantic air masses reaching Lampedusa may be more influenced by anthropogenic aerosol during their passage over Mediterranean Sea and continents.

The Smirnov et al. (2002) criterion has been widely used to identify marine aerosol type (e.g. Toledano et al., 2007; Pace et al., 2006). However, the threshold values for δ_a(500 nm) and α(440–870) proposed by Smirnov et al. (2002) to identify maritime aerosol type are purely empirical. Thus, the use of these thresholds could result in erroneous identification of this aerosol type. In fact, in Alboran Island we found measurements that fulfill this criterion but that are not associated with pure maritime conditions. In this sense, in Fig. 3 we show the δ_a(500 nm) and α(440–870) observed on 26 August 2002. During this day, the δ_a(500 nm) values ranged from 0.06 to 0.13 with mean daily value of 0.09 ± 0.01 and α(440–870) was in the range 0.3–0.6, indicating clean atmospheric condition dominated by coarse particles. Thus, according to the Smirnov et al. (2002) criteria (δ_a(500nm) < 0.15 and α(440–870) < 1) this day is classified as background maritime case. However, the back trajectory analysis (Fig. 3b) and MODIS satellite image (Fig. 3c) for 26 August revealed the presence of dust over Alborán Island. Therefore, care must be taken when using δ_a(500 nm) and α(440–870) thresholds for discriminating the background maritime cases since dusty situations with low dust loads can be confused with pure maritime conditions. Additional information such as air mass back trajectory or satellite images is needed for better identifying the background maritime cases.

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As can be seen in Fig. 2, there were several days strongly influenced by aerosols, with δ_a(500 nm) values exceeding 0.3. High aerosol loads (δ_a(500 nm) > 0.3) over Alborán Island were observed on 30 of the 160 analysed days. All these events were observed from July to October. In 27 of these cases, the mean daily α(440–870) values were lower than 0.8 and FMF lower than 0.5; suggesting predominance of coarse particles as either sea salt or dust transported from desert areas. According to the analyses of back trajectories and MODIS satellite images (not shown), all these 27 cases were related to dust intrusions from North Africa. It is important to note that in these dust events, the δ_F(500 nm) values were also relatively high (for this remote site) and ranged from 0.07 to 0.20 with mean value of 0.12 ± 0.03. These results highlight the considerable contribution of fine mode particles to the aerosol population (FMF ranged from 20 % to 52 %) during these dust events. Back trajectory analysis for days with dust intrusions with highest fine aerosol load revealed that the air masses reaching the study area at low levels (at 500 m or 1500 m level) have originated over northern Italy and the Mediterranean Sea. However, during desert dust events with lowest fine aerosol loads, none of the air masses affecting the study area come from northern Italy or Mediterranean Sea.

The remaining high aerosol load events were observed from 30 September to 4 October 2011 (Fig. 4). During these days the high aerosol loads were associated with relatively high α(440–870) values that reached the highest α(440–870) value (about 1.6) during the entire study on 4 October. During these days, the δ_F(500 nm) values were also high (> 0.19) and reached the highest mean daily value of 0.33 on 4 October. This behavior suggests the dominance of fine particles transported from continental industrial/urban areas as there are no local anthropogenic activities on the Island. The high $\delta_a(\lambda)$ observed in these cases were associated with persistent intense high pressure systems centered over the Azores, which favored transport of anthropogenic particles emitted from Europe to Alborán Island. Indeed, the back trajectory analysis revealed that these events were associated with air masses coming from Italy (Fig. 4c). Thus, the desert dust transport appears to be a main cause of high aerosol loads while trans-

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port from Italy is associated with occasional high aerosol loads over Alboran Island. These results are in accordance with those reported by Fotiadi et al. (2006) for Crete, who found the highest values of $\delta_a(\lambda)$ primarily during southeasterly winds, associated with coarse dust aerosols, and to a lesser extent to northwesterly winds associated with fine aerosols originated in urban industrial European areas.

3.2 Monthly variation of aerosol properties over Alborán Island

Figure 5a shows the monthly mean values of δ_a(500 nm), δ_F(500 nm), δ_C(500 nm) with the corresponding standard deviations for the analysed period. The monthly average data are calculated from daily averaged data. The largest values of δ_a(500 nm), reflecting high aerosol load, were observed during July–October while the lowest values (0.06–0.08) were measured from November to January. On other hand, the monthly mean values of α(440–870) and FMF were lower than 1.0 and 0.5 respectively, indicating a relatively high abundance of coarse particles in each month of the analysed period, except in October. For October, the mean α(440–870) was 1.1 ± 0.4 and the FMF 0.63 ± 0.20, indicating an increase in fine particle contribution during this month. It is also worth noting that both δ_F(500 nm) and δ_C(500 nm) showed a pronounced increases during July–October, suggesting increased loads of both fine and coarse particles during these months. Moreover, δ_C(500 nm) reached its maximum in August while δ_F(500 nm) peaked in October.

This pronounced change in aerosol loads from summer to winter in 2011 is primarily due to the seasonal change in atmospheric circulation over the Mediterranean (Fig. 5b). The increased coarse aerosol load observed during July–October was associated with the high frequency of desert dust intrusions in summer in comparison to November–January (Fig. 5b). In fact, 40 %, 70 %, 41 % and 14 % of measurement days in July, August, September and October were associated with Saharan dust intrusions while in November–January there were no Saharan dust intrusions (Fig. 5b). Moreover, the air mass recirculation over the western Mediterranean especially in summer (Millan et al., 1997) along with the increased photochemical activity due to the high insolation during this season may favor the accumulation of fine aerosols that can explain the high fine particle loads during July–October in comparison with November–January. In addition, the presence of these fine aerosol particles may be favored by pollution transport from Europe and coastal urban industrial areas in northeast Africa. In this sense, the highest fine mode aerosol optical depth observed in October was associated with the increase in the frequency of air masses coming from Italy (see for example Fig. 4). On other hand, the low aerosol loads registered in November–January were due to the high frequency of clean Atlantic air advection (70–100 %) and the absence of Saharan dust intrusions in this period (Fig. 5b). These results highlight the important role of the large scale circulation on monthly aerosol variation over Alborán Island.

3.3 Spatial variability of aerosol properties over western Mediterranean region

AERONET data of level 2 from Alborán and three AERONET stations surrounding the Island (see Fig. 1) obtained from July 2011 to January 2012 are considered in this study to investigate the spatial variation of aerosol optical properties over the western Mediterranean region. For analyzing the spatial aerosol variability we compared the aerosol data obtained over Alborán with those observed over these nearby sites using only time coincident measurements. In Table 2 average values of $\delta_a(\lambda)$, α(440–870), δ_F(500 nm) and FMF from July 2011 to January 2012 for Alborán, Málaga, Oujda and Palma de Mallorca are shown.

Temporal evolutions of daily mean values of δ_a(500 nm) from July 2011 to January 2012 obtained over Alborán Island and Málaga station are shown in Fig. 6. Daily mean data were calculated only from time coincident measurements for direct comparison. Málaga is located approximately 150 km northwest of Alborán. The temporal variations of daily mean values of δ_a(500 nm) were similar for both sites on most days of the analyzed period, indicating similarities in the processes that control the aerosol load over both sites. In fact, high correlation in δ_a(500 nm) with correlation coefficient, R, of 0.75 between these two sites was found. However, large differences are also present on some days (e.g. on 8 August 2011 at Alboran Island we registered

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δ_a(500 nm) above 0.5 while at Málaga the values were below 0.1). These differences are due, in large part, to the differences in the times of occurrence and intensity of Saharan dust intrusions over both sites. In fact, the correlation in δ_F(500 nm) between these sites, $R = 0.86$, was higher than the correlation in δ_C(500 nm), $R = 0.65$. Similar results were obtained when comparing the aerosol properties over Alborán with those in Oujda ($R = 0.82$ for δ_F(500 nm) and $R = 0.70$ for δ_C(500 nm)) and Palma de Mallorca ($R = 0.67$ for δ_F(500 nm) and $R = 0.32$ for δ_C(500 nm)).

For $\lambda > 500$ nm, values of $\delta_a(\lambda)$ were slightly larger over Alborán than over Málaga (Table 2). Indeed, the mean δ_a(1020 nm) value obtained at Alborán was 35 % larger than that observed over Málaga. This indicates that the coarse particles levels were significantly larger over Alborán in comparison with Málaga during the analysed period. In fact, the mean δ_C(500 nm) for the entire analysed period was slightly higher (0.09 ± 0.08) at Alborán in comparison with Málaga (0.06 ± 0.05). The lower coarse particles load over Málaga as compared to Alborán is likely due to the higher frequency of Saharan dust outbreaks over Alborán as compared to Málaga and also to dust deposition in its way from Alborán to Málaga. On other hand, for $\lambda < 500$ nm, the mean value of $\delta_a(\lambda)$ over Alborán was almost similar to that over Málaga (Table 2). It is interesting to note that the mean $\delta_F(\lambda)$ value for the entire studied period observed over Alborán (0.09 ± 0.06) was similar to that obtained (0.09 ± 0.06) over the Málaga urban coastal site, suggesting similar concentrations of fine particles over both sites. This result is almost surprising because Málaga is a coastal city with significant local anthropogenic emissions in comparison to Alborán where there are no local anthropogenic activities. As we commented before, Alborán Island is located near an important shipping route and hence it's expected to be highly influenced by ship emissions. Thus, these results suggest that emissions from ships may play in Alborán Island the role that traffic and urban activities play in Málaga.

A comparison of the $\delta_a(\lambda)$ obtained at Oujda and Alborán Island is shown in Table 2. Daily mean data were calculated only from time coincident measurements for direct comparison. In this case, the $\delta_a(\lambda)$ at all wavelengths were low at Alborán when

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compared to Oujda, indicating lower aerosol concentrations over Alborán. However, δ_F(500 nm) was similar over Oujda and Alborán (Table 2), indicating similar fine particle loads over both sites. This result is also surprising because Oujda is an urban site with significant local anthropogenic emissions in comparison to Alborán Island where there is no local anthropogenic activities. These results also point to the significant role that the ship emissions may play over Alborán. On the other hand, δ_C(500 nm) obtained over Oujda (0.14 ± 0.15) was higher than that observed over Alborán (0.11 ± 0.10), indicating higher coarse particle concentrations over Oujda. The large coarse particle load over Oujda may result from its proximity to dust sources and local dust re-suspension.

The mean $\delta_a(\lambda)$ values at all wavelengths over Alborán were much higher than those observed over Mallorca, especially at the larger wavelengths which are more influenced by coarse particles (Table 2). However, as in the other cases the δ_F(500 nm) was very similar over both sites (Table 2) in spite of the large distance (about 650 km) separating the sites and site characteristic differences. This result is also astonishing because Mallorca AERONET station is significantly affected by urban sources while Alborán Island is a remote site with no local anthropogenic activities. Thus, the observed decrease in δ_a(500 nm) from south (Alborán) to north (Mallorca) may be attributed to the proximity of Alborán Island to the dust sources in north Africa as compared to Mallorca. A gradient in dust load from south to north in western Mediterranean has also been reported by other authors (e.g., Barnaba and Gobbi, 2004). Overall, based on the above comparisons it may be concluded that the $\delta_C(\lambda)$ showed a south-to-north decrease in this region of western Mediterranean, while the fine mode aerosol optical depth was fairly similar over these sites.

3.4 Comparison between Maritime Aerosol Network and Alborán measurements

From July to November 2011 the Maritime Aerosol Network acquired measurements over the whole Mediterranean Sea, Black Sea and Atlantic Ocean from the ship Nautilius_11. Figure 7 shows δ_a(500 nm), δ_F(500 nm), δ_C(500 nm) and FMF obtained during

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this cruise. The measurements made over the Mediterranean Sea were divided, on the basis of the differences in the aerosol sources and air masses affecting each area, into three regions, western, central and eastern Mediterranean. As can be seen from Fig. 7, all the analysed aerosol properties showed large variability with no evident pattern during the cruise period. This large variability in aerosol properties during this cruise can be explained by the different aerosol sources and air masses that affected each region during the measurement period (see below). For the entire cruise period, the δ_a(500 nm) varied from 0.08 to 0.70 with a mean value of 0.22 ± 0.12, indicating a relatively large aerosol load over the cruise area. On the other hand, δ_F(500 nm) also showed large variability and ranged between 0.04 and 0.60 with a mean value of 0.16 ± 0.10 while δ_C(500 nm) fluctuated within the range 0.01–0.30 with mean value of 0.06 ± 0.04. For 85 % of the measurements, the fine mode fraction was in the range 0.52–0.96, indicating the predominance of situations dominated by fine mode particles during this cruise.

The highest δ_a(500 nm) values ranging from 0.20 to 0.46 with a mean value of 0.35 ± 0.09 were observed over the western Mediterranean Sea during the cruise period 28 September–8 October (Table 3). Also, δ_F(500 nm) values were highest (varying in the range of 0.14–0.40 with a mean value of 0.29 ± 0.09) over the western Mediterranean. These high aerosol loads were associated with high FMF values in the range 0.70–0.87, which show the predominance of fine anthropogenic particles over this area during this period (Fig. 7c). According to the back trajectory analyses, the air masses that affected the western Mediterranean region during this period come from the polluted areas of Italy which explains the observed large values of δ_a(500 nm) and the predominance of fine mode particles (see for example Fig. 4c). The aerosol loads were also relatively high over the Black Sea (δ_a(500 nm), ranging from 0.08 to 0.68 with a mean value of 0.25 ± 0.16 during 26 July–15 August cruise period) and were strongly dominated by fine particles as showed by FMF values ranging from 0.64 to 0.94. The large values of δ_a(500 nm) and those of δ_F(500 nm) (δ_F(500 nm) in the range 0.07–0.60) and the predominance of the fine mode over the Black Sea during this cruise

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period were associated, according to the HYSPLIT back trajectory analyses, with air masses coming from north-eastern Europe (figure not shown); this region has been identified as a strong source of pollutants in Europe (e.g., Barnaba and Gobbi, 2004). In contrast, the lowest δ_a(500 nm) values (varying in the range 0.08–0.26 with mean value of 0.14±0.06) were observed over eastern Mediterranean at the end of the cruise (5–13 November). These low δ_a(500 nm) values were associated with FMF ranging between 0.30 and 0.64, showing a predominance of coarse aerosol over this area during this period. It is worth noting that the aerosol loads over the eastern Mediterranean during 5–13 November decreased drastically in comparison with the aerosol levels observed in the same region during the cruise period from 18 August to 13 September (Table 3). The decrease was more pronounced for the fine particle load; δ_F(500 nm) fell from 0.16±0.07 in the first measurements over eastern Mediterranean to 0.07±0.02 in the last ones. In contrast, δ_C(500 nm) showed an increase from 0.04 ± 0.02 during 18 August–12 September to 0.08 ± 0.04 during 5–13 November. This drastic change may be explained by the seasonal changes in the meteorological conditions. In this sense, the last measurements over the eastern Mediterranean Sea were obtained during the end of autumn when aerosol wet deposition is more effective and secondary aerosol formation is less important than in summer, which may explain the lower aerosol loads observed at the end of the expedition.

It is worth noting that Fig. 7 has the disadvantage of providing the spatial variations of aerosol properties over the cruise area which implicitly includes the temporal variations of these properties. Thus, to analyze the spatial variation over the Mediterranean Basin during the cruise and avoid the inherent aerosol temporal variation we compared the aerosol data obtained over Alborán Island with those observed on board the ship Nautilius_11 using only time coincident measurements. For this, aerosol data obtained over Black Sea during 26 July–15 August and those observed over the central Mediterranean Sea during 13–28 September and 25 October–5 November as well as those obtained in the eastern Mediterranean Sea during 18 August–12 September and 5–13 November were compared to those observed over Alborán. Table 4 shows the

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comparison between the mean values of $\delta_a(\lambda)$, $\alpha(440–870)$, $\delta_F(500\,nm)$, $\delta_C(500\,nm)$ and FMF obtained at Alborán and those obtained over the Black Sea, central and eastern Mediterranean Sea regions during the cruise from July to November 2011. As can be seen from this Table, the mean $\delta_a(500\,nm)$ value obtained over Alborán was comparable to those measured over the Black Sea and central Mediterranean regions and significantly lower than that observed over the eastern Mediterranean region during the analyzed period. However, the mean $\delta_F(500\,nm)$ value obtained over Alborán was significantly lower than that measured over Black Sea, central and eastern Mediterranean regions, indicating that Alborán Island was significantly less influenced by anthropogenic particles during the cruise period compared to the other regions. In contrast, the mean $\delta_C(500\,nm)$ value obtained over Alborán island was higher than that measured over Black Sea, central and eastern Mediterranean regions indicating higher coarse particle loads over Alaborán compared to the other regions. The mean Angström exponent values observed over Black Sea, eastern and central Mediterranean regions were 1.7 ± 0.3, 1.7 ± 0.3 and 1.2 ± 0.4, respectively, while over Alborán, they were below 1.0, indicating the predominance of coarse particles over Alborán and the high dominance of fine anthropogenic particles over the other regions. According to HYSPLIT back trajectory analyses, the Black Sea, central and eastern Mediterranean regions were mostly affected by air masses coming from north-eastern Europe during the cruise over these areas while Alborán Island was mostly affected by air masses originated from North Africa (figures not shown). This can explains the predominance of coarse particles and the high coarse particle loads observed over Alborán and the predominance of fine particles over the Black Sea, central and eastern Mediterranean regions during the cruise over these areas.

3.5 Evaluation of the impacts of the European ship emission regulations on the atmospheric columnar aerosol properties

For evaluating the impact of the European ship emission regulations on columnar aerosol properties over the western Mediterranean region, continuous sun photometer

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measurements from the Málaga AERONET station (operating since November 2008) during 2009–2012 are analyzed here. The station is located at approximately 4 km from the port and thus is expected to be largely influenced by emissions from the ships and port due to sea/land breeze circulation effects. Figure 8a and b shows box and whisker plots of δ_a(500 nm) and α(440–870) daily values measured at Málaga in 2009, 2010, 2011 and 2012. As can be seen in Fig. 8a, the annual mean and median $\delta_a(\lambda)$ values were similar in 2009, 2010, 2011 and 2012, indicating no significant change in the annual aerosol load. In addition, the 25 % and 75 % percentiles were also similar in 2009, 2010, 2011 and 2012, suggesting no significant changes in the background and high aerosol loads. Also, the annual mean, median and 25 % and 75 % percentiles of α(440–870) values do not show any significant changes, suggesting no significant change in aerosol types over the site study during the analysed period. Furthermore, the ANOVA test at a significant level of 0.05 shows that there were no significant statistical differences in $\delta_a(\lambda)$ and α(440–870) measured in 2009, 2010, 2011 and 2012. As we saw before, the study area is frequently influenced by Saharan dust intrusions from North Africa and hence the inter-annual variation in the desert dust intensity and frequency can mask the desired objective (reduction of pollutant levels) of the European directive on emissions from maritime transport. To avoid Saharan dust interference, we analyzed the δ_F(500 nm) which is mostly associated with fine anthropogenic particles. Figure 7c shows box and whisker plots of δ_F(500 nm) values measured at Málaga in 2009, 2010, 2011 and 2012. The mean annual values of δ_F(500 nm) were similar (about 0.08 ± 0.05), indicating no significant impact of the European directive on fine aerosol loads over the study area. Furthermore, the 25 % and 75 % percentiles in these years do not show clear differences indicating no significant changes in the background and peak loads. In summer the Mediterranean Basin is characterized by atmospheric stagnation and sea-land breezes, and consequently, shipping emissions can affect a large area. So to augment the chance for detecting the possible impact of European directive we performed the same previous analysis but using only data obtained in the summer months during 2009–2012. However, the results are similar to those obtained previously; thus suggesting no pronounced change in aerosol properties over Málaga since the Directive was implemented in 2010.

The relationship between pollutant emissions and their levels in the atmosphere is not straightforward. Atmospheric aerosol levels in specific areas are affected not only by local emissions, but also by long-range transport, natural emissions and meteorological conditions. So, the absence of change in the aerosol load over Málaga AERONET station may be due to drastic changes in meteorological and synoptic conditions or to increases in the pollutant emissions from other local anthropogenic activities or other Mediterranean countries. Further studies using columnar aerosol measurements at various Mediterranean sites are needed to evaluate the impact of this European Directive on columnar atmospheric aerosols over the Mediterranean Sea.

4 Conclusions

AERONET sun photometer measurements obtained over Alborán Island and three nearby different sites in the western Mediterranean were analyzed in order to investigate the temporal and spatial variations of columnar aerosol properties over this poorly explored region. Furthermore, these data were also evaluated for identifying the cause of high aerosol load events and evaluating the possible impact of EU ship emission regulations on aerosol properties over the western Mediterranean.

Within the analysed period the daily average values of δ_a(500 nm) over Alborán Island ranged from 0.03 to 0.54 with a mean and standard deviation for the entire period of 0.17 ± 0.12, indicating high aerosol load variation. The observed mean δ_a(500 nm) value over Alborán Island was significantly higher than the reported for open oceanic areas not affected by long range aerosol transport (0.06–0.08). The α(440–870) values were lower than 1 for 70 % of the measurement days, suggesting that coarse particles dominated the aerosol population over the Alborán Island for the majority of the measurement days.

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High aerosol loads over Alborán were mainly associated with desert dust transport from arid areas in North Africa and occasional advection of anthropogenic fine particles from Italy. The aerosol optical depth values of fine mode during dust events were also relatively high (for this remote site), suggesting that the fine mode particles also have considerable influence on optical properties during these dust events. Background maritime conditions over Alborán were observed on about 40 % of the measurement days during the analyzed period; almost all of these days were characterized by advection of clean Atlantic air masses over the study area.

The mean value of δ_F(500 nm) over Alborán Island was comparable to that observed over the other three nearby AERONET stations, suggesting homogeneous spatial distribution of fine particle loads over the four studied sites in spite of the large differences in local sources. South to north decreases in $\delta_C(\lambda)$ was found which was probably associated with increased desert dust deposition from south to north or decreased dust frequency from south to north.

Aerosol properties acquired on board the ship Nautilius_11 within Maritime Aerosol Network over the whole Mediterranean Sea, Black Sea and Atlantic Ocean from July to November 2011 showed large variability with no evident pattern. In 85 % of the measurements, the fine mode fraction was in the range 0.52–0.96, indicating the predominance of fine mode particles over the cruise areas during the monitoring period. The highest δ_a(500 nm) and δ_F(500 nm) mean values of 0.35 ± 0.09 and 0.29 ± 0.09 during the cruise period were observed over the western Mediterranean Sea, which were related to polluted air masses coming from northern Italy. In contrast, the lowest δ_a(500 nm) values (mean value of 0.14 ± 0.06) during this cruise were observed over the eastern Mediterranean Sea on the final days of the cruise in autumn, when aerosol wet deposition is more effective and secondary aerosol formation is less important than in summer.

The mean δ_a(500 nm) value obtained over Alborán Island was comparable to those measured on board the ship Nautilius_11 over the Black Sea and central Mediterranean regions, and significantly lower than those observed over the eastern Mediter-

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ranean. However, the mean δ_F(500 nm) values obtained over Alborán were significantly lower than those measured over the Black Sea and central and eastern Mediterranean regions, indicating that Alborán Island was significantly less influenced by anthropogenic particles than the other regions. The Black Sea, central and eastern Mediterranean regions were mostly affected by air masses coming from Europe during the cruise over these region, explaining the high predominance of fine particle over these regions. In contrast, Alborán Island was mostly affected by advection from North Africa which explains the predominance of coarse particles and the high coarse particle loads observed over this site.

Finally, the annual mean, median and 25 % and 75 % percentiles of $\delta_a(\lambda)$, δ_a(500 nm) and α(440–870) values in 2009, 2010, 2011 and 2012 registered over Málaga port city, which is expected to be more influenced by ship emissions, do not show any significant changes, suggesting that there were no significant change in columnar aerosol properties related to implementation of European Directive in 2010 in the southwestern Mediterranean region. The absence of change in the columnar aerosol load over Málaga AERONET station may be due to drastic changes in meteorological and synoptic conditions or to increases in the pollutant emissions from other local anthropogenic activities or other Mediterranean countries. Further studies using columnar aerosol measurements at various Mediterranean sites are needed to evaluate the impact of this European Directive on atmospheric aerosols over the Mediterranean Sea.

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Table 1. Statistical summary of daily mean values of spectral aerosol optical depth at 1020, 500 and 340 nm, Angström exponent, α(440–870), fine and coarse mode aerosol optical depths at 500 nm, δ_F(500 nm) and δ_C(500 nm), and fine mode fraction, FMF, observed over Alborán Island in the western Mediterranean during July 2011–January 2012; SD is the standard deviation and COV is the coefficient of variation.

	Mean	SD	Minimum	Maximum	COV(%)
δ_a(1020 nm)	0.11	0.10	0.01	0.46	91
δ_a(500 nm)	0.17	0.12	0.03	0.54	70
δ_a(340 nm)	0.25	0.15	0.05	0.65	60
α(440–870)	0.8	0.4	0.2	1.7	50
δ_F(500 nm)	0.08	0.05	0.01	0.30	63
δ_C(500 nm)	0.10	0.09	0.01	0.4	90
FMF	0.47	0.15	0.20	0.94	32

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Table 2. Average values and standard deviations of $\delta_a(\lambda)$, α(440–870), δ_F(500 nm) and FMF from July 2011 to January 2012 for Alborán Island, Málaga, Oujda and Palma de Mallorca. Only days with coincident measurements at Alborán and at each one of the additional AERONET stations are used for direct comparison.

	Alborán	Málaga	Alborán	Palma de Mallorca	Alborán	Oujda
δ_a(1020 nm)	0.09 ± 0.09	0.06 ± 0.05	0.13 ± 0.10	0.06 ± 0.04	0.13 ± 0.11	0.16 ± 0.17
δ_a(870 nm)	0.10 ± 0.09	0.08 ± 0.06	0.14 ± 0.11	0.08 ± 0.05	0.14 ± 0.11	0.18 ± 0.18
δ_a(670 nm)	0.12 ± 0.10	0.09 ± 0.07	0.16 ± 0.12	0.10 ± 0.06	0.16 ± 0.12	0.19 ± 0.18
δ_a(500 nm)	0.16 ± 0.11	0.14 ± 0.09	0.20 ± 0.13	0.14 ± 0.07	0.20 ± 0.13	0.23 ± 0.19
δ_a(440 nm)	0.18 ± 0.12	0.16 ± 0.10	0.23 ± 0.14	0.18 ± 0.09	0.22 ± 0.14	0.25 ± 0.19
δ_a(380 nm)	0.21 ± 0.13	0.20 ± 0.12	0.26 ± 0.15	0.21 ± 0.10	0.25 ± 0.15	0.29 ± 0.20
δ_a(340 nm)	0.23 ± 0.14	0.23 ± 0.13	0.29 ± 0.16	0.24 ± 0.11	0.28 ± 0.16	0.30 ± 0.20
α(440–870)	0.9 ± 0.4	1.0 ± 0.3	0.8 ± 0.4	1.2 ± 0.4	0.8 ± 0.4	0.8 ± 0.4
δ_F(500 nm)	0.09 ± 0.06	0.09 ± 0.06	0.09 ± 0.07	0.09 ± 0.06	0.09 ± 0.07	0.09 ± 0.06
FMF	0.50 ± 0.15	0.53 ± 0.13	0.47 ± 0.18	0.60 ± 0.14	0.47 ± 0.19	0.47 ± 0.19
Number of coincident days	141	141	93	93	101	101

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Table 3. Mean values of δ_a(500 nm), δ_F(500 nm), δ_C(500 nm), α(440–870) and FMF obtained over Black Sea, western, central and eastern Mediterranean Sea and Atlantic Ocean during the Nautilius ship cruise from July to November 2011.

Region	δ_a(500 nm)	δ_F(500 nm)	δ_C(500 nm)	α(440–870)	FMF
Black Sea (26 Jul–15 Aug)	0.25 ± 0.16	0.21 ± 0.14	0.04 ± 0.03	1.76 ± 0.30	0.82 ± 0.08
Eastern Mediterranean I (18 Aug–12 Sep)	0.20 ± 0.08	0.16 ± 0.07	0.04 ± 0.02	1.74 ± 0.20	0.81 ± 0.08
Central Mediterranean I (13–28 Sep)	0.18 ± 010	0.12 ± 0.09	0.06 ± 0.03	1.27 ± 0.40	0.66 ± 0.08
Western Mediterranean (28 Sep–8 Oct)	0.35 ± 0.09	0.29 ± 0.09	0.07 ± 0.02	1.50 ± 0.13	0.80 ± 0.07
Atlantic Ocean (9–19 Oct)	0.19 ± 0.10	0.11 ± 0.05	0.09 ± 0.06	1.08 ± 0.25	0.56 ± 0.09
Central Mediterranean II (25 Oct–5 Nov)	0.22 ± 0.10	0.13 ± 0.07	0.09 ± 0.04	1.05 ± 0.30	0.57 ± 0.13
Eastern Mediterranean II (5–13 Nov)	0.14 ± 0.06	0.07 ± 0.02	0.08 ± 0.04	0.90 ± 0.35	0.49 ± 0.12

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Table 4. Comparison between the mean values of aerosol properties obtained at Alborán Island and those obtained over Black Sea, central and eastern Mediterranean Sea regions during ship cruise from July to November 2011, using only time coincident measurements.

	Alborán	Black Sea	Alborán	Central Mediterranean	Alborán	Eastern Mediterranean
δ_a(440 nm)	0.29 ± 0.16	0.33 ± 0.20	0.23 ± 0.11	0.25 ± 0.12	0.11 ± 0.07	0.24 ± 0.10
δ_a(500 nm)	0.26 ± 0.18	0.27 ± 0.17	0.21 ± 0.09	0.20 ± 0.10	0.10 ± 0.06	0.19 ± 0.08
δ_a(675 nm)	0.24 ± 0.17	0.16 ± 0.11	0.16 ± 0.07	0.14 ± 0.07	0.07 ± 0.05	0.12 ± 0.05
δ_a(870 nm)	0.22 ± 0.17	0.11 ± 0.08	0.14 ± 0.0.6	0.11 ± 0.05	0.05 ± 0.04	0.08 ± 0.03
α(440–870)	0.5 ± 0.4	1.7 ± 0.3	0.7 ± 0.3	1.2 ± 0.4	1.0 ± 0.4	1.7 ± 0.3
δ_F(500 nm)	0.08 ± 0.04	0.16 ± 0.10	0.09 ± 0.05	0.13 ± 0.08	0.05 ± 0.03	0.14 ± 0.07
δ_C(500 nm)	0.18 ± 0.14	0.06 ± 0.04	0.10 ± 0.05	0.07 ± 0.03	0.05 ± 0.05	0.04 ± 0.03
FMF	0.37 ± 0.17	0.70 ± 0.16	0.45 ± 0.11	0.62 ± 0.13	0.60 ± 0.16	0.75 ± 0.15
Number of coincident measurements	26	26	22	22	47	47

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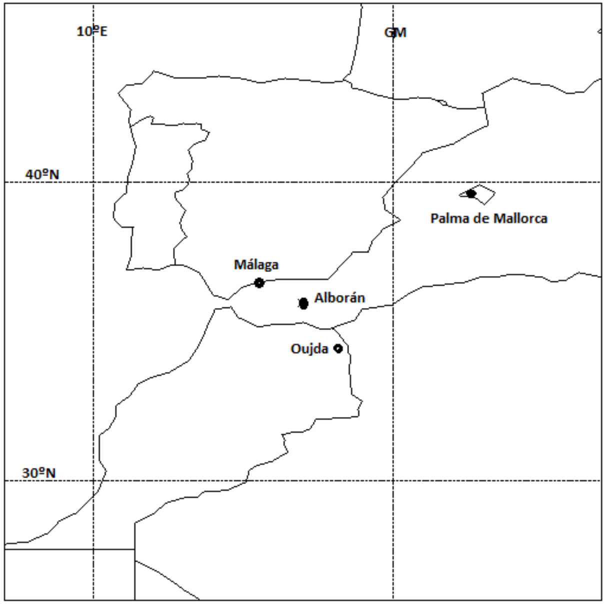

Figure 1. Map of the western Mediterranean showing the location of Alborán Island, Málaga, Oujda and Palma de Mallorca.

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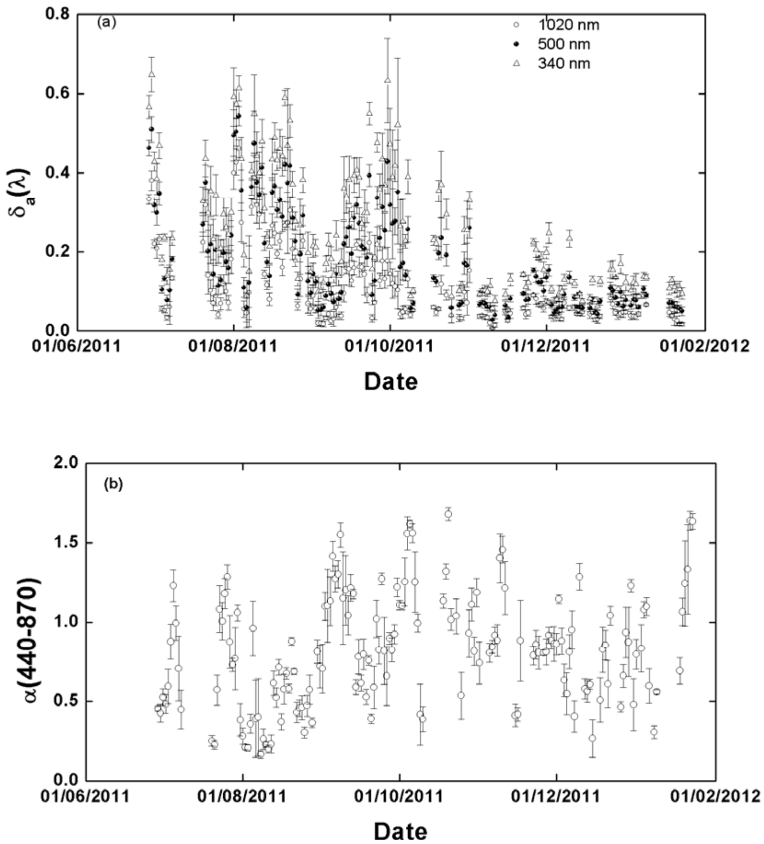

Figure 2. Temporal evolution of the daily mean values of **(a)** aerosol optical depth at 340, 500 and 1020 nm and **(b)** the Angström exponent calculated in the range 440–870 nm, measured at Alborán Island in the western Mediterranean from July 2011 to January 2012. The error bars are standard deviations.

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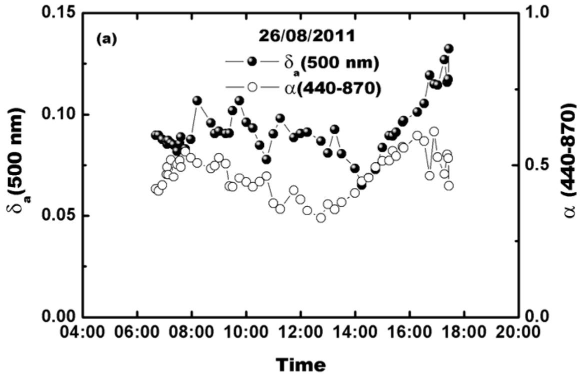

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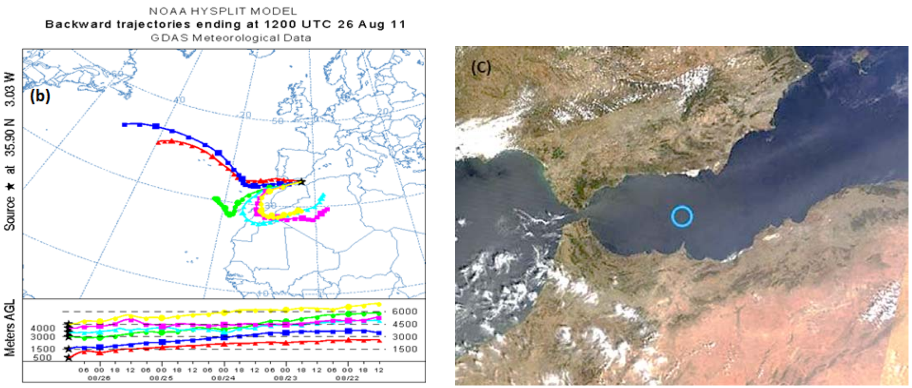

Figure 3. (a) Aerosol optical depth at 500 nm and Angstrom exponent in the range 440–870 nm, **(b)** backward trajectories ending at 12:00 UTC over Alborán Island at height altitudes of 500, 1500, 3000 and 4000 m and **(C)** MODIS satellite image for 26 August 2011.

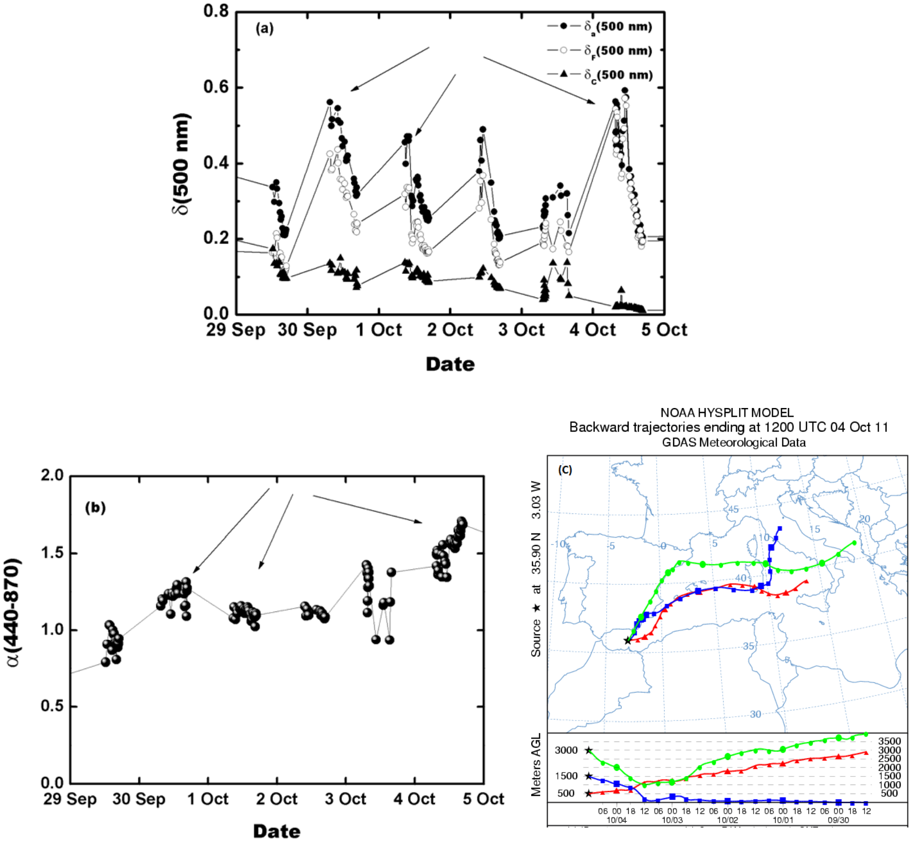

Figure 4. (a) Total, fine and coarse aerosol optical depths at 500 nm and **(b)** Angstrom exponent in the range 440–870 nm obtained at Alborán Island during 29 September–5 October 2011. **(c)** Backward trajectories ending at 12:00 UTC on 4 October 2011 over Alboran Island at altitudes of 500, 1500, 3000 m.

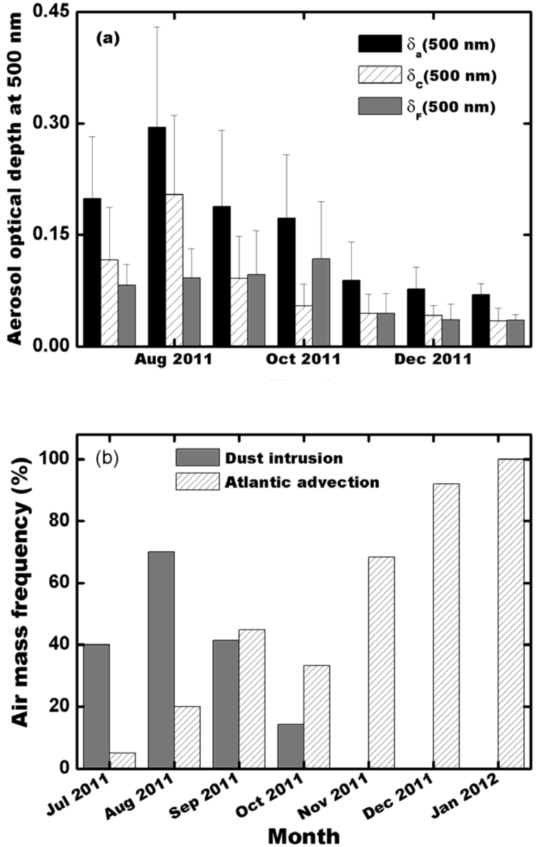

Figure 5. (a) Monthly variations of total, coarse and fine mode optical depths at 500 nm obtained at Alborán Island from July 2011 to January 2012. The error bars are standard deviations. **(b)** Monthly frequency of Saharan dust intrusions and Atlantic air mass advections over Alborán Island from July 2011 to January 2012.

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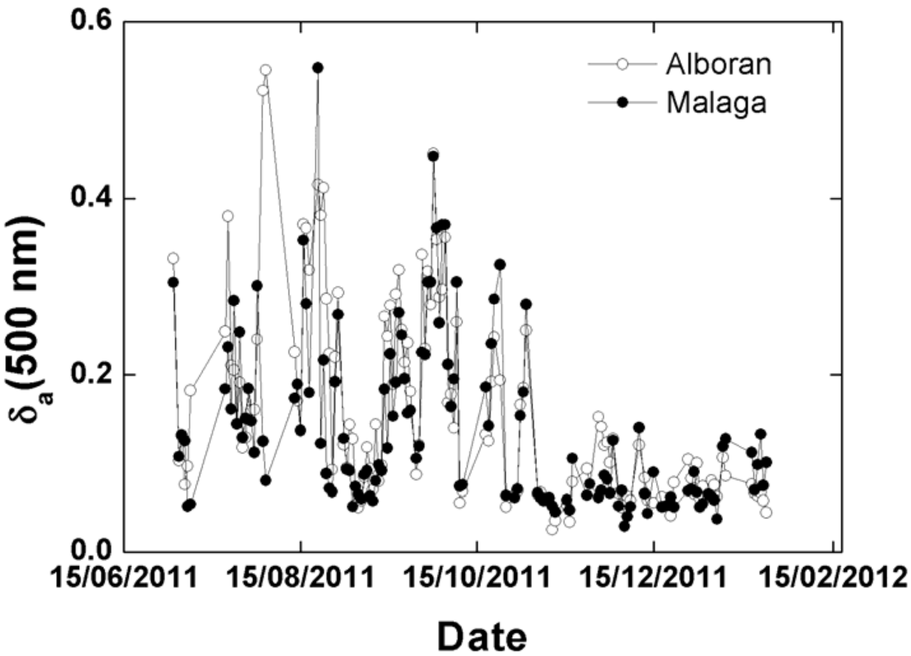

Figure 6. Temporal evolutions of daily mean values of δ_a(500 nm) from July 2011 to January 2012 obtained over Alborán Island and Málaga. Daily mean data were calculated only from time coincident measurements.

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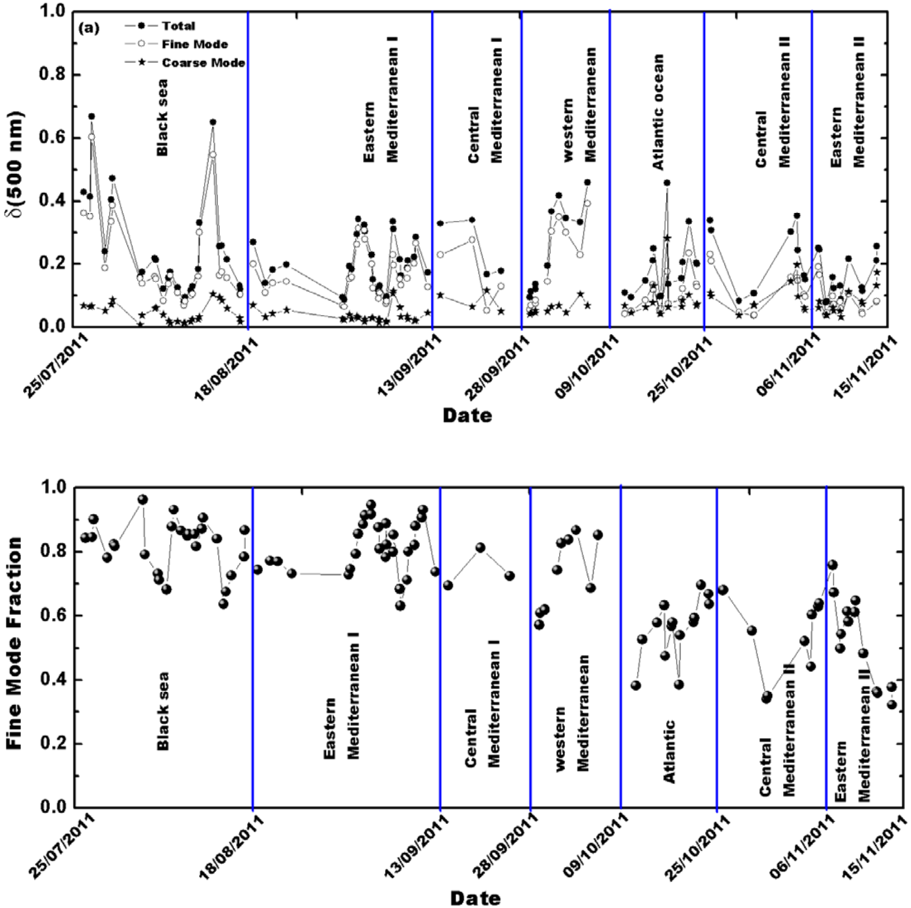

Figure 7. Temporal evolutions of δ_a(500 nm), δ_F(500 nm), δ_C(500 nm), and FMF obtained on board of Nautilius ship. The data belong to the Maritime Aerosol Network (MAN) and were acquired between July and November 2011.

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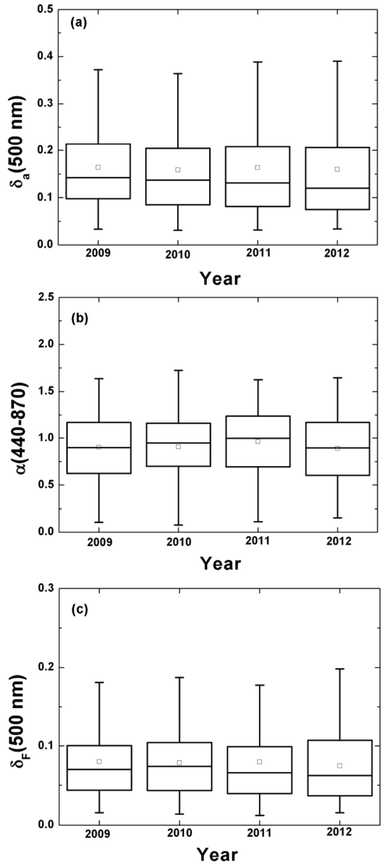

Figure 8. Box and whisker plots of daily average values of **(a)** of $\delta_a(\lambda)$, **(b)** $\alpha(440\text{–}870)$ and **(c)** $\delta_F(500\,\text{nm})$ obtained at Málaga in 2009, 2010, 2011 and 2012. Line within box: median value; square symbol: mean value; top and bottom of box: third and first quartile.

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