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African dust outbreaks over the Mediterranean Basin during 2001–2011: PM_{10} concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology

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Abstract. The occurrence of African dust outbreaks over the whole Mediterranean Basin has been studied on an 11-yr period (2001–2011). In order to evaluate the impact of such mineral dust outbreaks on ambient concentrations of particulate matter, PM_{10} data from regional and suburban background sites across the Mediterranean area were compiled. After identifying the daily influence of African dust, a methodology for the estimation of the natural dust contributions on daily PM_{10} concentrations was applied.

Our findings point out that African dust outbreaks are sensibly more frequent in southern sites across the Mediterranean, from 30 to 37 % of the annual days, whereas they occur less than 20 % of the annual days in northern sites. The central Mediterranean emerges as a transitional area, with slightly higher frequency of dust episodes in its lower extreme when compared to similar latitudinal positions in western and eastern sides of the Basin. A decreasing south to north gradient of African dust contribution to PM_{10}, driven by the latitudinal position of the monitoring sites at least 25° E westwards across the Basin, is patent across the Mediterranean. As a result of this, an experimental equation for the estimation of annual African dust contributions based on the latitudinal position was obtained. From 25° E eastwards, higher annual dust contributions are encountered due to the elevated annual occurrence of severe episodes of dust but also because of inputs from Negev and Middle Eastern deserts. The slightly higher frequency of African dust episodes observed over southern sites in the central Mediterranean Basin is compensated by its moderately lower intensity. Concerning seasonality patterns and intensity characteristics, a clear summer prevalence is observed in the western part, with low occurrence of severe episodes (daily dust averages over 100 $\mu g\,m^{-3}$ in PM_{10}); no seasonal trend is detected in the central region, with moderate-intensity episodes; and significantly higher contributions are common in autumn-spring in the eastern side, with occurrence of various severe episodes throughout the year. Overall, African dust emerges as the largest PM_{10} source in regional background southern sites of the Mediterranean (35–50 % of PM_{10}), with seasonal peak contributions to PM_{10} up to 80 % of the total mass.

The multi-year study of African dust episodes and their contributions to PM_{10} concentrations reveals a consistent decreasing trend in the period 2006/2007 to 2011 in 4 of the 17 studied regions, all of them located in the NW of the Mediterranean. Such decrease is almost parallel to that of the NAO (North Atlantic Oscillation) index for the summer period, progressively more negative since 2006. Therefore, a sharp change in the atmospheric circulation over the last 5 yr (a similar negative NAO period occurred in the 1950 decade) have affected the number of African dust episodes and consequently the annual dust inputs to PM_{10} observed in the NW part of the Mediterranean. By investigating mean temperatures and geopotential height maps at 850 hPa it is evident a displacement of warm air masses accomplishing African dust towards the central Mediterranean in the 2007–2008 biennium, and towards the NW African coast and the Canary Islands in the 2009–2011 triennium.

1 Introduction

On a global scale, most of the atmospheric particles are emitted by natural sources, mineral dust being the second more abundant component after sea-spray derived aerosols (IPCC, 2007). These crustal aerosols are mainly released to the atmosphere from arid and semiarid regions located in subtropical areas in the Northern Hemisphere, with the Sahara-Sahel-Chad dust corridor being the largest source region in north Africa (Prospero et al., 2002; Moreno et al., 2006).

In general, atmospheric circulation over north-western Africa is mainly controlled by the northeast trade winds and by the mid-tropospheric Saharan Air Layer. Southerly of the Saharan deserts, winds are usually mono-directional with a general westward transport. Along the year, the dust plume extension varies accordingly to the displacement of the Inter-Tropical Convergence Zone (ITCZ) (Prospero et al., 1981). In winter, when the ITCZ is at its southernmost position, dust originated in Sahara and Sahel deserts is transported towards the tropical Atlantic Ocean by such northeast trade winds (Alonso-Pérez et al., 2011). In summer, such trade winds are more constrained owing to the northern displacement of the ITCZ (Prospero et al., 1981). Additionally, high insolation and temperatures over Sahara-Sahel area create strong surface winds and large-scale convection processes, which lift dust particles at high atmospheric levels (up to 5 km). Such mineral dust particles, transported at high-altitude by the Saharan air layer, move partially towards the tropical Atlantic above the boundary layer (Bergametti et al., 1989a). A significant amount of the atmospheric dust is transported also towards the western Mediterranean along an anticyclonic gyre over north-western Africa. Regardless of most of the African dust particles are exported westwards over the Atlantic (Viana et al., 2002; Alastuey et al., 2005; Alonso-Pérez et al., 2011), travelling for long distances and impacting very distant areas in the Caribbean and the United States (Arimoto et al., 1997; Prospero et al., 2002), a considerable amount of dust is also released northerly, affecting the Mediterranean region (Ganor and Mamane, 1982; Bergametti, et al., 1989b; Guerzoni and Chester, 1996; Querol et al., 1998; Rodriguez et al., 2001; Escudero et al., 2005, Gerasopoulos et al., 2006; Kallos et al., 2006; Koçak et al., 2007; Mitsakou et al., 2008; Papadimas et al., 2008) and even other European areas (Klein et al., 2010).ç

African dust mobilization may be studied from ground-based measurements (which is important to evaluate air quality, health outcomes, ecosystem damages, visibility reduction), or from a wider perspective by studying the atmospheric column (which is necessary to evaluate climatic feedbacks). Ground-based and columnar measurements are not necessarily correlated. From a ground-based monitoring perspective, African dust towards the Mediterranean region is usually mobilized by a number of meteorological scenarios widely described elsewhere (Rodriguez et al., 2001; Escudero et al., 2005; Gkikas et al., 2009, 2012). As summarized in Querol et al. (2009a), dust-storms affecting western and central Mediterranean are caused by low-pressure systems over the Atlantic or north Africa, high pressures over the Mediterranean, or high pressures at upper levels over NW Africa. Dust storms over the eastern Mediterranean are generally originated by cyclones moving eastwards throughout the Mediterranean, but also because of the combination of low pressures over north Africa with high pressures over Middle East. Details on these meteorological scenarios may be found in Escudero et al. (2005) for the western and central scenarios and in Kallos et al. (2006) for the eastern ones.

One of the most important health outcomes of African dust concerns its chemical and biological composition, as highlighted in a recent review study (Karanasiou et al., 2012). The meteorological scenarios favouring the export of African dust to the western or eastern Mediterranean imply that mineral particles emitted in regions located in east Africa, such as the Bodele Depression, hardly reach western areas in the Mediterranean. Conversely, dusts from NW deserts usually affect eastern locations of the Mediterranean because of the sweep effect caused by low-pressures movement. Taking into account that significant differences in natural soil composition are observed from one region to another in north Africa (Moreno et al., 2006), variations in anthropogenic pollutants travelling with mineral dust are also observed (Perrino et al., 2010; Rodríguez et al., 2011) and the content in micro-organisms may be different, the potential effects on health (Pérez et al., 2008; Polymenakou et al., 2008; Tobías et al., 2011a), ecosystems (Arimoto, 2001) and climate (IPCC, 2007; Papadimos et al., 2012) may vary notably.

Yearly, variations in mean ambient temperature or rainfall amount are observed. These changes are associated to alterations in the atmospheric circulation. Thus, it is expected that such variations in the atmospheric dynamics affect also other phenomena such as dust mobilization frequency and/or intensity. The study of long data series of dust contributions at multiple points across a wide area may be indicative of periodic or consistent tendencies. As an example, Cusack et al. (2012) have observed a clear decreasing trend in a number of components of $PM_{2.5}$ at a regional background site in NE Spain linked to the implantation of abatement measures at regional and continental scales, but also associated to meteorological cycles. In fact, the general decrease in PM levels observed at ground-based monitoring sites north to south in western and central Europe (Barmpadimos et al., 2012; Cusack et al., 2012) is essentially attributed to recent, and probably cyclic, changes in the atmospheric circulation over the northern Atlantic.

Bearing in mind that African dust is an important source of particulate matter pollution in specific areas, causing by itself or contributing to exceed the daily limit values of PM_{10} (in Europe being established in $50\,\mu g\,m^{-3}$ by the 2008/50/EC Directive), and exerting negative health outcomes (Middleton et al., 2008; Mitsakou et al., 2008; Pérez et al., 2008; Jiménez et al., 2010; Mallone et al., 2011; Zauli Sajani et al., 2011; Samoli et al., 2011a, b; Tobías et al., 2011a, b), the identification of such episodes and the quantification of daily and annual contributions of desert dust to PM is currently necessary. Moreover, the identification of temporal trends in African dust contributions at ground-based monitoring sites may be indicative of atmospheric circulation changes.

Previously, Querol et al. (2009a) performed a comprehensive work on African dust significance over the Mediterranean from a database of up to 6 yr at 21 sites. In the present work, a much longer database (based on ground-based measurements) of PM concentrations and African dust contributions (up to 11 yr) at multiple locations (17 areas with at least one monitoring site in each one) across the Mediterranean has been interpreted. The aim of this work is to characterize the phenomenology of African dust outbreaks across the Mediterranean, with special interest in identifying the significance of this natural PM source on ambient air PM_{10} concentrations by studying daily and seasonal patterns. Moreover, an extended database of African dust episodes across the whole Mediterranean together with their contributions to PM_{10} allows a confident study on inter-annual trends.

This study is part of the LIFE Programme European project MED-PARTICLES (Particles size and composition in Mediterranean countries: geographical variability and short-term health effects). Overall the MED-PARTICLES project aims at quantifying short-term health effects of particulate matter over the Mediterranean region by distinguishing different particle sizes, chemical components and sources, with special emphasis in the effects of African dust. Since the main motivation of the project is in evaluating health effects, the results of this study are crucial from an epidemiological point of view. These results will be used to estimate effects on health distinguishing between PM_{10} from north Africa and that from local and/or regional sources.

2 Methodology

2.1 Data collection

To evaluate African dust contributions across the Mediterranean Basin (not including north African sites) and to assess their impact on PM_{10} levels, data from 19 regional background (RB) and sub-urban background (SUB) sites were obtained (Fig. 1): 7 in Spain covering central and eastern Iberia, and the Balearic islands; 2 in southern France; 5 in Italy covering north to south the peninsula, Sardinia and Sicily; 1 in Bulgaria; 3 in Greece, being 2 in the continent and 1 in Crete; and 1 in Cyprus. Thus, these ground-based monitoring sites are distributed west to east and north to south of the Mediterranean region, with less spatial coverage of the area located 25° E eastwards, as shown in Fig. 1. From all these regional background sites daily PM_{10} concentrations have been obtained from 2001 to 2011 when available. All the data used in this study were obtained from public European databases: Airbase (http://acm.eionet.europa.eu/databases/airbase/), EMEP (www.emep.int/) and EUSAAR (http://www.eusaar.net/).

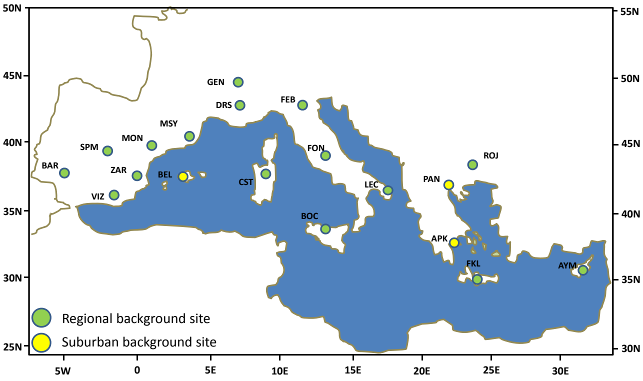

Fig. 1. Location of regional and suburban background sites providing data for this study.

Station characteristics, coordinates and altitude above sea level, data coverage, and measurement principles are shown in Table 1. Among the 19 ground-based monitoring sites, 3 different techniques have been used to determine PM_{10} concentrations: gravimetric determinations at the Spanish EMEP sites, at Montseny, Finokalia and Ayia Marina; and real time monitors (BETA and TEOM) with different measurement principles (Beta gauche attenuation and oscillating microbalance, respectively) at the rest of monitoring sites. Real time concentrations should be corrected against gravimetric ones. Since only official data (transferred from the countries after different validation steps to the European Environmental Agency, EMEP programme and EUSAAR/ACTRIS databases) are used in this work, data quality should be guaranteed.

2.2 African dust occurrence

The methodology used for identifying African dust episodes is the same as in previous studies (Rodríguez et al., 2001; Escudero et al., 2005, 2007; Querol et al., 2009a; Pey et al., 2010). This procedure assures the identification of almost all the African dust episodes, independently of their intensity, and consists in the interpretation of a couple of tools: meteorological products (NCEP/NCAR : http://www.esrl.noaa.gov/psd/data/composites/hour/), aerosol maps (BSC-DREAM: http://www.bsc.es/projects/earthscience/DREAM/; NAAPS-NRL: http://www.nrlmry.navy.mil/aerosol/; SKIRON: http://forecast.uoa.gr/dustindx.php), satellite images (SeaWiFS: http://oceancolor.gsfc.nasa.gov/SeaWiFS/; MODIS: http://modis.gsfc.nasa.gov/), and air masses back-trajectories (HYSPLIT: http://ready.arl.noaa.gov/HYSPLIT_traj.php). Firstly, back-trajectories of air masses are interpreted to account for the transport of air masses from north Africa and

Table 1. Location, type (regional background: RB; sub-urban background: SUB), data availability and measurement methods used in the monitoring sites of this study.

Country	Code	Type	Latitude	Longitude	Altitude (m a.s.l.)	Start	End	Method
SPAIN	MSY	RB	41°45′36″ N	02°35′00″ E	728	2002	2011	GRAV
	MON	RB	40°56′48″ N	00°17′27″ W	570	2001	2011	BETA
	ZAR	RB	39°05′10″ N	01°06′07″ W	885	2001	2011	GRAV
	VIZ	RB	37°14′18″ N	03°28′28″ W	1265	2001	2011	GRAV
	BAR	RB	38°28′33″ N	06°55′22″ W	393	2001	2011	GRAV
	SPM	RB	39°31′29″ N	04°21′09″ W	1241	2001	2011	GRAV
	BEL	SUB	39°33′50″ N	02°37′22″ E	117	2001	2011	BETA
FRANCE	GEN	RB	45°43′55″ N	04°58′50″ E	235	2001	2010	TEOM
	DRS	RB	44°31′15″ N	05°05′24″ E	460	2004	2010	TEOM
ITALY	FEB	RB	44°45′04″ N	10°26′05″ E	1020	2005	2010	BETA
	FON	RB	41°40′48″ N	13°40′48″ E	393	2001	2010	BETA
	LEC	RB	40°27′32″ N	18°06′58″ E	10	2009	2010	BETA
	CST	RB	39°03′52″ N	08°27′26″ E	270	2005	2010	BETA
	BOC	SUB	38°07′13″ N	13°18′07″ E	141	2001	2010	BETA
GREECE	PAN	SUB	40°35′20″ N	23°01′54″ E	363	2001	2010	BETA
	APK	SUB	37°59′36″ N	23°49′10″ E	290	2001	2010	BETA
	FKL	RB	35°20′00″ N	25°40′00″ E	150	2004	2010	GRAV
BULGARIA	ROJ	RB	41°41′45″ N	24°44′19″ E	1750	2005	2010	BETA
CYPRUS	AYM	RB	35°02′21″ N	33°03′29″ E	532	2003	2010	GRAV

MSY: Montseny; MON: Monagrega; ZAR: Zarra; VIZ: Víznar; BAR: Barcarrota; SPM: San Pablo de los Montes; BEL: Castillo de Bellver; GEN: Genas; DRS: Drôme Rurale Sud; FEB: Febbio; FON: Fontechiari; LEC: Lecce; CST: Censt; BOC: Boccadifalco; PAN: Panorama; APK: Agia Pareskevi; FKL: Finokalia; ROJ: Rojen Peak; AYM: Ayia Marina.
GRAV: Gravimetric; BETA: Beta Attenuation monitor; TEOM: Tapered Element Oscillating Microbalance.

Negev-Middle Eastern deserts. Secondly, aerosol maps and satellite images are evaluated, which usually result in the consideration of additional days impacted by dust. Finally, for specific cases in which some doubts arise, meteorological maps are calculated to verify the existence of favourable scenarios for the transport of dust. It is important to remark that, in some cases when dust air masses are travelling at high altitude, African dust may affect PM levels at ground levels up to 2 days after the episode ends, as discussed in the European Guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe (http://ec.europa.eu/environment/air/quality/legislation/pdf/sec_2011_0208.pdf). Thus, a final evaluation of PM levels at the different RB and SUB sites is conducted, which incorporated these possible delays.

2.3 African dust contribution to PM_{10} concentrations

In order to ascertain on daily African dust contributions to PM_{10} and $PM_{2.5}$ concentrations, a statistical methodology applied to the PM data series have been used. This method is based on the application of 30 days moving 40th percentile to the PM_{10} or $PM_{2.5}$ data series, after excluding those days impacted by African dust. For those days affected by African dust it is obtained a percentile value which is assumed to be the theoretical background concentration of PM if African dust didn't occur. After that, the African dust contribution is obtained by difference between the experimental PM_{10} or $PM_{2.5}$ concentration value and the calculated 40th percentile value.

This methodology was initially published (Escudero et al., 2007) considering the 30th percentile. Subsequently this method was slightly modified by adopting the 40th percentile instead the 30th one only for conservative reasons. Currently, this is one of the official methods adopted by the European Commission for evaluating the occurrence of African dust outbreaks and quantifying its contributions (Commission staff working paper establishing guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe). It is important to remark that the feasibility of this method was demonstrated by comparing experimentally measured concentrations of mineral matter determined at three Spanish RB sites versus the estimated African dust contributions obtained by this procedure.

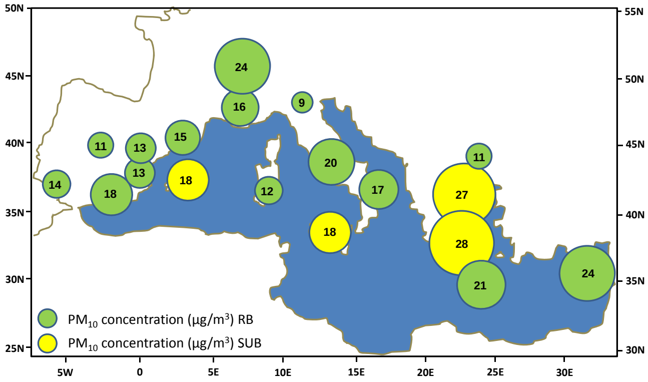

Fig. 2. Annual PM_{10} levels ($\mu g\,m^{-3}$) at regional (RB) and suburban (SUB) background sites across the Mediterranean for the period 2001–2011.

3 Results and discussions

3.1 PM levels across the Mediterranean

Annual averages of PM_{10} at RB sites across the Mediterranean Basin (note that monitoring sites in north Africa were not studied) reflect a wide spatial variability (Fig. 2), with the highest concentrations in eastern-basin areas (21–24 $\mu g\,m^{-3}$), but also nearby Lyon (Genas, 24 $\mu g\,m^{-3}$). On the contrary, the lowest PM_{10} concentrations (9–11 $\mu g\,m^{-3}$) are observed at high-altitude sites west to east of the Mediterranean: San Pablo de los Montes, EMEP site in central Spain at 1241 m a.s.l.; Febbio, RB site in northern Italy at 1020 m a.s.l.; and Rojen Peak, EMEP site in the Rhodopes (Bulgaria) at 1750 m a.s.l. Intermediate PM_{10} concentrations are recorded in the rest of RB sites, being sensibly higher in the vicinity of densely populated and/or industrialized areas, and to north Africa. This is the case of: Fontechiari, in close proximity to Rome (20 $\mu g\,m^{-3}$); Víznar, near Granada and north Africa (18 $\mu g\,m^{-3}$); Lecce, in southern Italy (17 $\mu g\,m^{-3}$); Drome Rurale Sud, close to the highly industrialized area of Marseille (16 $\mu g\,m^{-3}$); and Montseny, in the vicinity of the Barcelona metropolitan and industrial agglomeration (15 $\mu g\,m^{-3}$). Concerning average PM_{10} concentrations at the suburban environments used in this study (selected because of the lack of RB sites covering that geographical areas), they are much higher in the Athens and Thessalonica influence areas (27–28 $\mu g\,m^{-3}$) than in Sicily or Majorca (18 $\mu g\,m^{-3}$). See Fig. A1 to appreciate average PM_{10} levels without the influence of African dust contributions.

Overall, there is an increasing PM gradient (occasionally broken as in the vicinity of Barcelona, Marseille and Rome, because of the high influence of anthropogenic emissions) from the NW to the SE of the Mediterranean. This augment coincides partially with that of African dust, but is mainly driven by the increase of the regional pollution towards the eastern part of the Basin. This increment was formerly reported and chemically characterized by Querol et al. (2009a, b). In those studies, they found higher concentrations of sulphate and carbonaceous aerosols easterly in the Basin, both components with a prevalent anthropogenic origin at these areas.

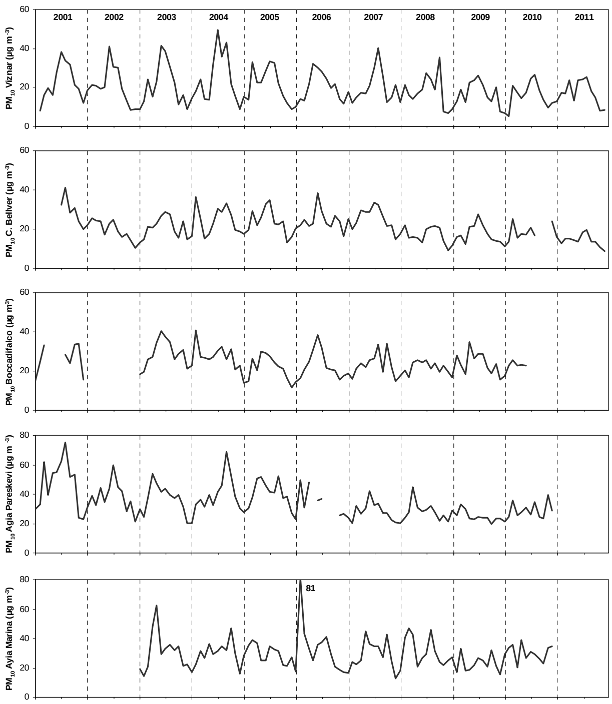

Fig. 3. Monthly PM_{10} levels ($\mu g\,m^{-3}$) at selected regional and suburban background sites across the Mediterranean Basin in the period 2001–2011: Víznar (SE Spain); C. Bellver (Balearic Islands); Boccadifalco (Sicily); Agia Pareskevi (Athens); Ayia Marina (Cyprus).

3.2 PM levels across the Mediterranean: seasonal patterns

PM levels show clear seasonal patterns across the Mediterranean (Fig. 3). In general, a summer maximum is observed throughout the basin attributed to a number of factors including: (1) stagnant conditions over the region and occurrence of a more stable planetary boundary layer over the sea in summer (Dayan et al., 1989; Pey et al., 2009); (2) enhanced formation of secondary pollutants owing to intense solar radiation and humidity; (3) high frequency of wildfires in Mediterranean and surrounding areas; (4) increased anthropogenic pressure since the Mediterranean is a common tourist destination; (5) reduced precipitation and aerosol wet removal; (6) higher emissions from maritime traffic (cruises and small boats) during the warm season. Specifically in the western side of the basin, the highest frequency of Saharan dust outbreaks (Querol et al., 1998; Rodríguez et al., 2001) and the effect of recirculation of air masses over that area (Millán et al., 1997; Rodríguez et al., 2002; Pey et al., 2009) contribute to increase the background levels. Likewise, the transport of polluted air masses from eastern Europe accounts for such summer increase in the eastern part (Gerasopoulos et al., 2006; Koulouri et al., 2008). Similarly at both sides, the lowest PM concentrations are observed in December-January, coinciding with well-ventilated conditions, low photochemical activity, higher precipitation amounts and less frequency of Saharan dust episodes. The most noticeable difference between western and eastern sides corresponds to the late winter-early spring period, when PM concentrations in the eastern Mediterranean are at their maximum due to the impact of severe African dust outbreaks, whereas they are low or intermediate in the western part.

3.3 African dust outbreaks: frequency

Figure 4a shows the average frequency of African dust outbreaks across the Mediterranean Basin during the period 2001–2010. It is evident the decreasing gradient of African dust outbreaks frequency from south to north of the Mediterranean Basin. Among the investigated areas, the lowest frequencies of African dust events are observed in central and NE Spain, SE France and northern Italy (17–18 %). On the contrary, the highest frequency is recorded in Sicily (37 %), followed by Cyprus (34 %, affected also by dust outbreaks from Negev and Middle Eastern deserts). As a result, a similar frequency of African dust is observed in south-western and eastern Iberia (23–24 %) and in central Italy and northern Greece (24 %). Furthermore, there is a linear relation between mean frequency of African dust outbreaks and latitude (Fig. 4b). The linear relation is almost the same for areas located in the eastern or western part of the Mediterranean, whereas it is slightly different for the central part of the Basin, where slightly higher frequency of dust episodes is observed in southern sites with respect to similar latitudinal points at both extremes of the Basin. This fact is directly related with the phenomenology of African dust episodes. As reviewed in Querol et al. (2009a), the western Mediterranean is more affected by African dust in summer. In contrast, the eastern Mediterranean is frequently impacted by African dust air masses in the autumn–spring period. Summer episodes usually affect the central part of the Mediterranean, especially from central Italy towards the south. Similarly, autumn–spring episodes impact often south Italy and Sicily. Thus, the central Mediterranean may be considered as a transitional area in terms of dust outbreaks phenomenology, which is impacted in its lower part by African dust all over the year. An additional issue concerns the African dust export towards the Mediterranean. From a ground-based monitoring perspective, the transport of African dust towards the western Mediterranean mostly occurs at relatively high atmospheric levels, exported from NW African deserts (Escudero et al., 2005). Over the eastern Mediterranean, however, most of the African dust affecting ground-based measurements is transported commonly at surface levels (Querol et al., 2009a). These features influence the intensity of the African dust outbreaks at both sides of the basin, as shown in following sections.

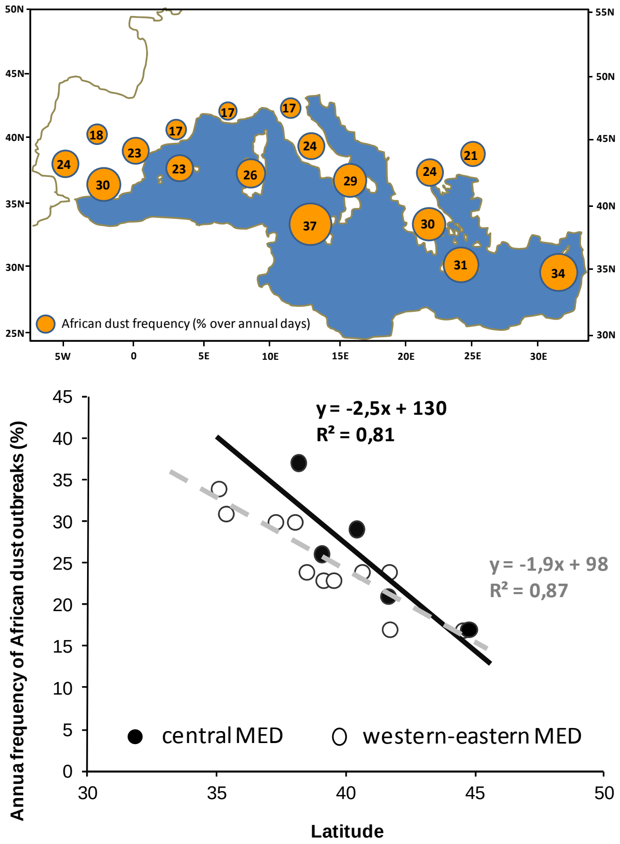

Fig. 4. (a) Top: mean frequency of African dust outbreaks (%) across the Mediterranean Basin during the period 2001–2011; **(b)** bottom: mean frequency of African dust outbreaks (%) versus latitude across the Mediterranean Basin during the period 2001–2011.

3.4 African dust outbreaks contributions to ambient air PM_{10}

3.4.1 Average PM_{10} concentrations

After identifying daily occurrence of dust outbreaks, the methodology described in Sect. 2.3 has been applied in order to estimate the dust contributions to PM_{10}. Figure 5 shows the average African dust contributions to the mean ambient air PM_{10} levels calculated for the existing periods (in most cases a 10–11 yr database was available). As seen in Fig. 5a, African dust inputs are considerably higher in the eastern locations of the Mediterranean when compared to those observed in the western side. In general, average annual concentrations were found to be maximum in Cyprus (8.4 $\mu g\,m^{-3}$) and Crete (7.3 $\mu g\,m^{-3}$), and minimum in NE Spain, SE France and north Italy (1.1–1.5 $\mu g\,m^{-3}$). The slightly higher dust impact (+1 $\mu g\,m^{-3}$) observed in Cyprus with respect to that of Crete might be explained by

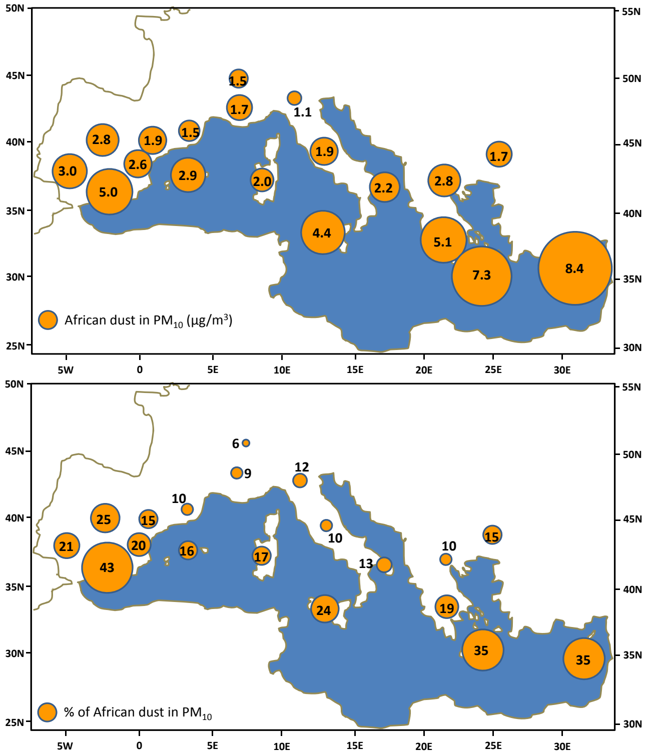

Fig. 5. (a) Top: mean African dust contributions to PM_{10} (in $\mu g\,m^{-3}$) across the Mediterranean (average values for the periods when data are available, in most cases from 2001–2010); **(b)** bottom: percentage of African dust over bulk PM_{10} registered in the monitoring sites selected in this study (average values for the periods when data are available, in most cases from 2001–2010).

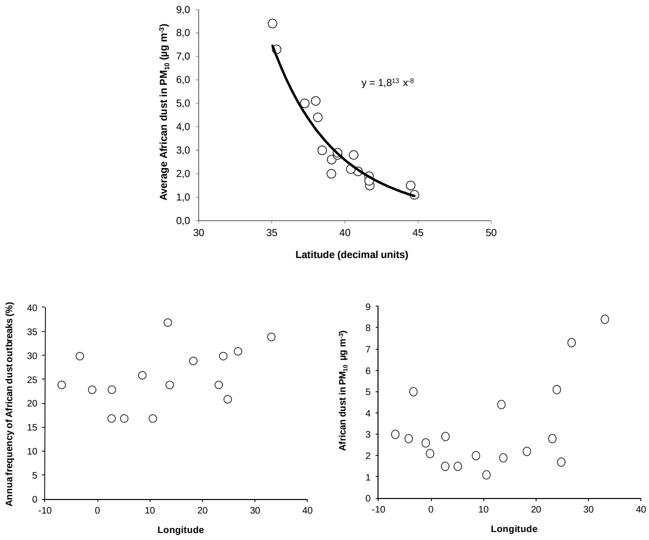

Fig. 6. (a) Top-left: mean annual African dust contributions in PM_{10} (in $\mu g\,m^{-3}$) versus latitude; **(b)** bottom-left: mean annual frequency of African dust outbreaks (%) versus longitude; **(c)** bottom-right: mean annual African dust contributions in PM_{10} (in $\mu g\,m^{-3}$) versus longitude.

the contribution of Negev and Middle Eastern deserts. In relative proportions with respect to the total PM_{10} (Fig. 5b), African dust is a dominant component in SE Spain, Crete and Cyprus (35–43 % of the PM_{10}), abundant in SW and central Spain, Sardinia and south Greece (19–25 % of the PM_{10}), and less important northerly and/or close to highly populated areas such as Barcelona, Marseille-Lyon, Rome and Thessaloniki (6–10 % of the PM_{10}).

In order to evaluate the factors governing such spatial distribution, a crossover study between average African contributions versus latitude and longitude has been conducted (Fig. 6a and b). As seen in Fig. 6a, there is a clear dependence of African dust contribution in PM_{10} with latitude, comparable to that observed during a severe African dust episode over the Iberian Peninsula (Cabello et al., 2012). It is remarkable that this relation is not linear but exponential, being defined by the following experimental Eq. (1)

$$y = 1.8^{13} x^{-8} \tag{1}$$

where y is the estimated African dust (in $\mu g\,m^{-3}$), and x is latitude (in decimal units).

Thus, for given latitude across the Mediterranean Basin, the expected average African dust contribution may be calculated by applying this experimental equation. This approach may be relevant in terms of air quality since allows a quick estimation of the contribution of African dust to PM concentrations on an annual basis. In the European context, this approach could be a preliminary step to be adopted by air quality managers in different countries affected by dust outbreaks (but currently not considering such episodes) before implementing some routinely method to justify such natural inputs.

Complementarily, the effect of the longitudinal position with respect to the average frequency of African dust outbreaks has been evaluated (Fig. 6b). The derived relationship is valid either for the regions where stations are found or near them. Despite that slight differences have been found between the position within the Basin and the frequency of African dust (Fig. 4b, Fig. 6b), locations situated at the same latitude and different longitude across the Mediterranean register on average similar African dust concentrations (Fig. 6c) if they are between 10° W and 25° E. At more eastern longitudes ($> 25°$ E, only two ground-based stations) it is evident a higher African dust contribution (Fig. 6c), having in mind that the frequency of African dust outbreaks does not increment. Regardless of the low number of monitoring sites, such observation is strongly related with the severity of some African dust episodes in the eastern part of the Basin, as mentioned in following sections but also with the contribution of dust from Negev and Middle Eastern deserts (Derimian et al., 2006; Basart et al., 2009) in the case of Cyprus.

Overall, mean annual African dust contributions in PM_{10} varied with respect to the latitude. A longitudinal effect is

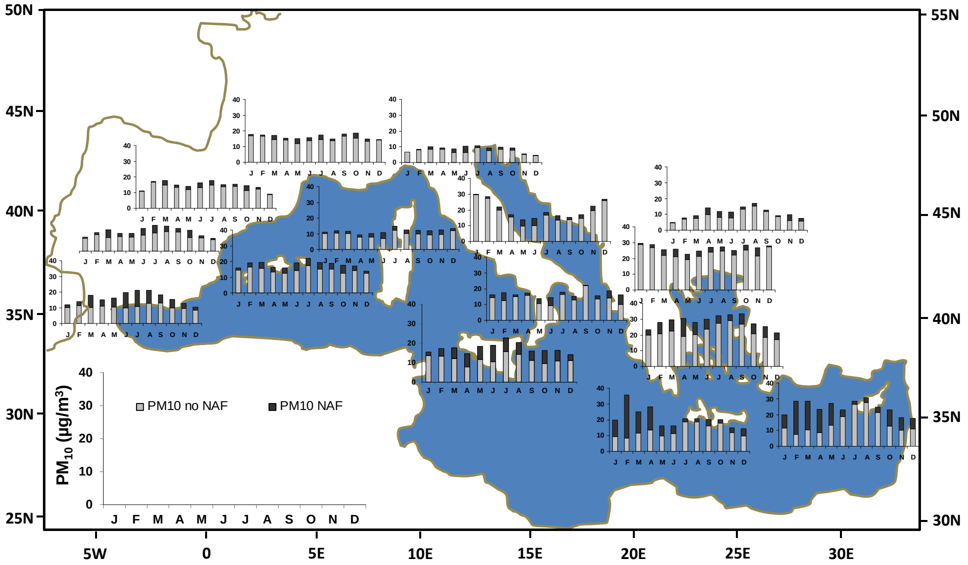

Fig. 7. Seasonal partitioning of PM_{10} (in $\mu g\,m^{-3}$), considering the influence of African dust (average values for the periods were data are available, in most cases from 2001–2010) across the Mediterranean Basin. NAF: African dust outbreaks.

patent from 25° E eastwards. The decreasing gradient of African dust towards the north is not lineal but exponential.

3.4.2 Seasonal patterns

As mentioned previously, African dust transport occurs in different seasons in western and eastern sides of the Mediterranean. These seasonal patterns exert a clear influence in the African dust contributions along the year.

Figure 7 shows the seasonal contributions of African dust on PM_{10}, and the partitioning between bulk ambient air PM_{10} and African dust. On average, African dust may occur all along the year across the Mediterranean. However, African dust inputs in the western side of the Mediterranean are considerably higher between May and October, and in March, when compared to the rest of the year. On the contrary, such inputs are clearly higher between November and May in the eastern part of the Mediterranean. An intermediate outcome is observed for central locations in the Mediterranean, where only slightly higher summer contributions are detected.

In relative terms, in the south-western part of the Mediterranean, African dust may account for about 50 % of PM_{10} mass in mid summer, and less than 10 % in winter. Similarly, in the most southern site of the central Mediterranean Basin, between 35 and 50 % of the PM_{10} in summer is mineral dust from north African deserts. More evident is the impact of the African dust in the most south-eastern locations studied, where up to 80 % of the PM_{10} recorded in the period February–April is constituted by African dust. By contrast, mineral dust from desert regions account for less than 10 % of PM_{10} in mid summer. The mentioned trends are repeated in the northern areas across the basin, although a substantial diminution in the dust contributions on ambient PM_{10} concentrations is evident.

3.4.3 Intensity of dust outbreaks

One of the most interesting aspects of African dust episodes concerns their intensity. By analyzing extended databases of African dust events across the Mediterranean it is possible to identify the occurrence and recurrence of intense African dust episodes. Figure 8 represents the percentage of African dust days according to their intensity (mean contributions in $\mu g\,m^{-3}$ in 9 concentration ranges, the highest being 30–44, 44–99 and > 100) for the whole Mediterranean. The occurrence of extreme dust events (with daily PM_{10} African dust contributions higher than $100\,\mu g\,m^{-3}$) with respect to the total number of episodes is infrequent. However, the occurrence of such severe outbreaks is relatively frequent (2–5 % of the African dust days) in the most south-eastern sites of this study (Greek Islands, south of Greece and Cyprus). As a result, some of these events occur randomly every year. The occurrence of extreme events is unusual (from less than 1 to 1 % of the African dust days) at equivalent latitudes westerly of the Mediterranean (southern Spain, southern Italy, Sicily). Moderate to intense events (30–99 $\mu g\,m^{-3}$ PM_{10} dust) are observed both in western, central and eastern areas, accounting for 15 to 25 % of the dust events in southern areas, 10–15 % in intermediate latitudes, and 5–10 % in northern areas. Low-intensity episodes (1–10 $\mu g\,m^{-3}$) prevail in northern locations of the western and central Mediterranean, accounting for 50 to 70 % of the African dust episodes.

Overall, the intensity characteristics described in this section are strongly related with the transport patterns of African

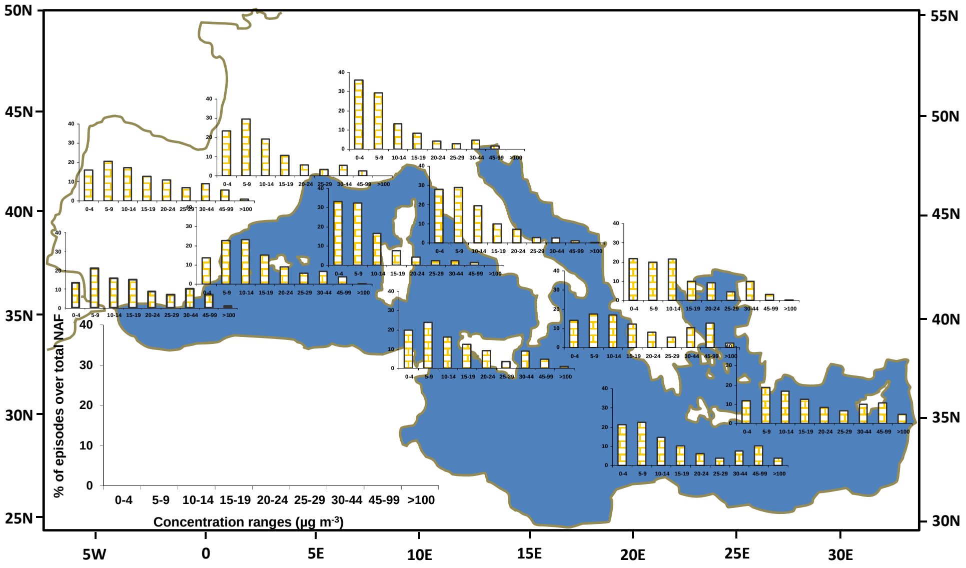

Fig. 8. Percentage of African dust episodes according to their intensity (divided in 9 concentration intervals from 0–4 to $> 100\,\mu\text{g}\,\text{m}^{-3}$) in selected areas across the Mediterranean Basin.

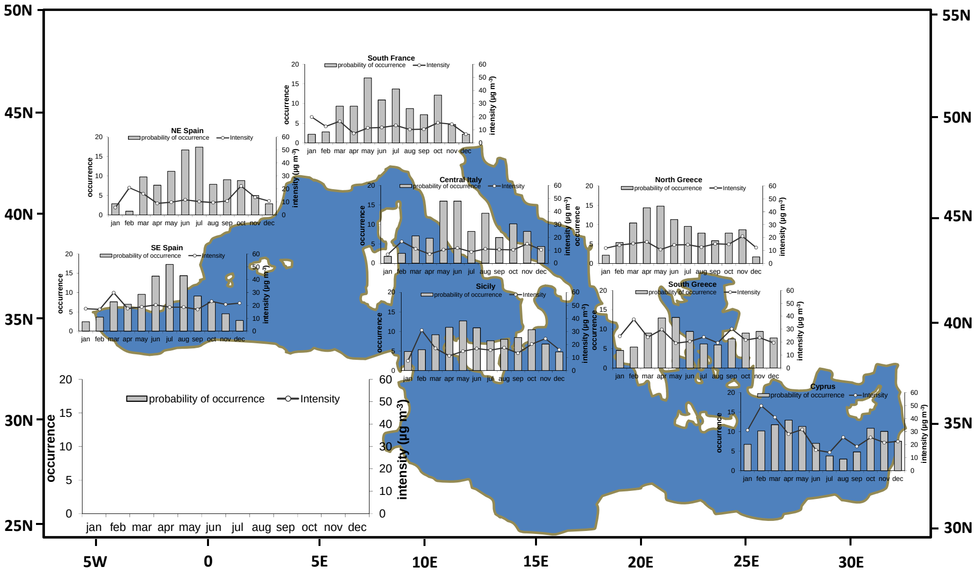

Fig. 9. Relation between probability of occurrence of African dust episodes (grey bars, in %) and intensity (black line, in $\mu\text{g}\,\text{m}^{-3}$) in selected areas across the Mediterranean Basin.

air masses. Dust transfer over western and eastern sides of the Mediterranean is caused by different transport mechanisms. In this context, the Atlas mountainous barrier, with a 2500 km extension from western Sahara towards Tunisia and peak altitudes up to more than 4000 m a.s.l., plays a dominant role in local and mesoscale atmospheric circulation patterns. As a result, African dust episodes over the western and central part of the Mediterranean are very frequent in summer (Rodríguez et al., 2001; Escudero et al., 2005), although moderate in intensity given the intricate transport processes (dust is travelling at high altitudes). The situation in the eastern part of the Mediterranean is significantly different. African dust transport is typically induced by cyclones moving eastwards across the Mediterranean and/or north Africa, transporting dust at surface levels. These flows may be enhanced during specific scenarios (Moulin et al., 1998) if air masses are canalized southerly of the Atlas Mountains (with a north-eastern prevalent direction), giving to the occurrence of short but intense dust episodes in the eastern side of the Mediterranean. From autumn to spring, when

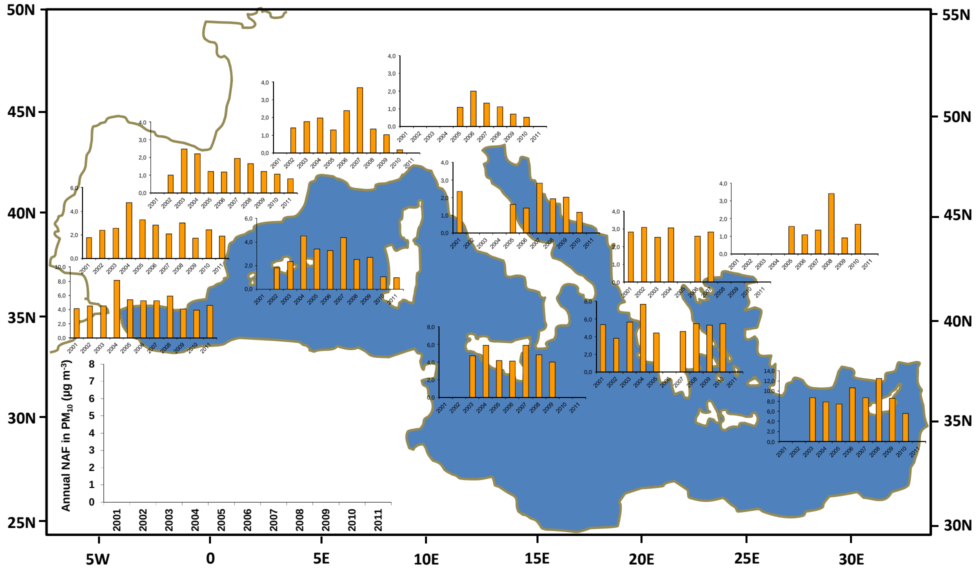

Fig. 10. Mean annual African dust contributions to PM_{10} (in $\mu g\,m^{-3}$) across the Mediterranean from 2001 to 2011 (when available). Note that different Y-axis scales are presented in the map.

dust episodes are more frequent over the eastern Mediterranean, dust plumes leaving north African deserts eastwards of Tunisia do not reach elevated atmospheric layers (Kalivitis et al., 2007; Nastos, 2012) since they do not find any significant geographical barrier neither convective processes are intense over north Africa. On the contrary, African dust air masses affecting the eastern Mediterranean in summer are encountered at high altitudes, due to the strong vertical convective flows over the dust source regions (Gobbi et al., 2000; Kalivitis et al., 2007; Papayannis et al., 2008).

In order to assess on the relation between African dust occurrence and intensity, Fig. 9 has been elaborated. Most of the patterns observed in Fig. 9 have been already discussed previously. Nevertheless, there are new specific features to be described and interpreted. In general, there is no clear relationship between occurrence and intensity, with significant seasonal variation of dust episodes but flat variation in the intensity along the year. Only in the eastern side of the Mediterranean, intensity and occurrence are varying in parallel. Concerning the intensity of dust occurrences, although limited variation is observed, February–March and October–November episodes over the western and central Mediterranean are usually more intense than those of other months. This fact is due to the transport mechanisms of the dust, always at ground level, and induced by two main scenarios already described in Escudero et al. (2005): (1) cyclone systems moving from western Iberia towards the Mediterranean; (2) widespread anticyclone over the western and central Mediterranean that moves slightly easterly. Commonly, the first scenario is typical of autumn, whereas the second is frequently observed in late autumn and early spring.

3.5 African dust outbreaks: inter-annual variability

Long-term series of annual averages of African dust contributions are helpful to recognize cyclic or climatic tendencies, which would be related to fast changes in atmospheric circulation patterns. Also variations in the emissions from source areas could be recognized by exploring these long-term databases. Figure 10 summarizes the annual averages of African dust contributions from 2001 to 2011 for selected areas across the Mediterranean. Although it was not possible to find a very complete PM_{10} database for the whole Mediterranean to quantify the African dust impact, usual data coverage ranges between 7 to 11 yr.

In general over central and southern areas (central and south Spain, central Italy and Sicily, Greece, and Cyprus), there are no evident increasing neither decreasing trends of African dust contribution to ambient PM_{10}, with sporadic annual peaks as a result of the occurrence of severe episodes. Thus, south Spain in 2004 registered a number of episodes above 150 $\mu g\,m^{-3}$; Sicily also recorded a very intense event in February 2004, with a daily concentration of more than 300 $\mu g\,m^{-3}$ of African dust; south Greece experienced also severe African dust episodes in February, May and November 2004, with daily peak concentrations up to 200 $\mu g\,m^{-3}$; Cyprus suffered numerous and very intense dust episodes in February–March, August and December 2008, 12 days with dust daily concentrations exceeding 100 $\mu g\,m^{-3}$.

The situation in the northern locations of the western and central Mediterranean (NE Spain, the Balearic Islands, SE France, and north Italy) is clearly different. In these areas, a prominent decreasing trend on ambient PM_{10} contributions is patent from 2006 or 2007 onwards. Before 2007, alternate

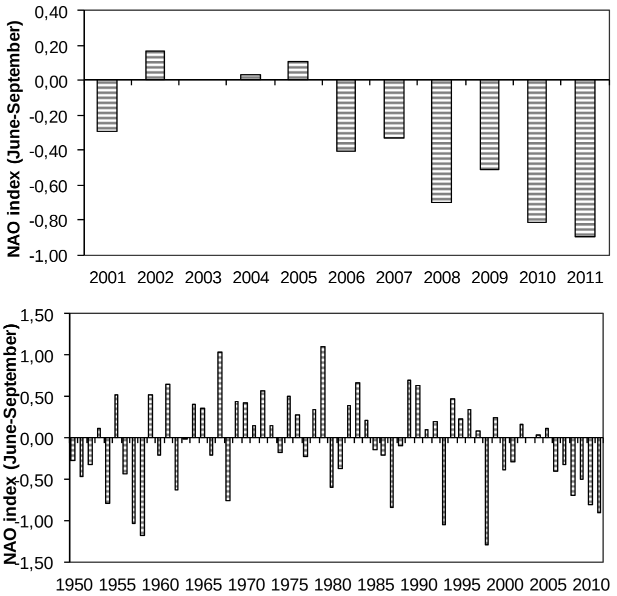

Fig. 11. (a) Top: NAO index calculated for the period June–September from 2001 to 2011; **(b)** bottom: NAO index calculated for the period June–September from 1950 to 2011.

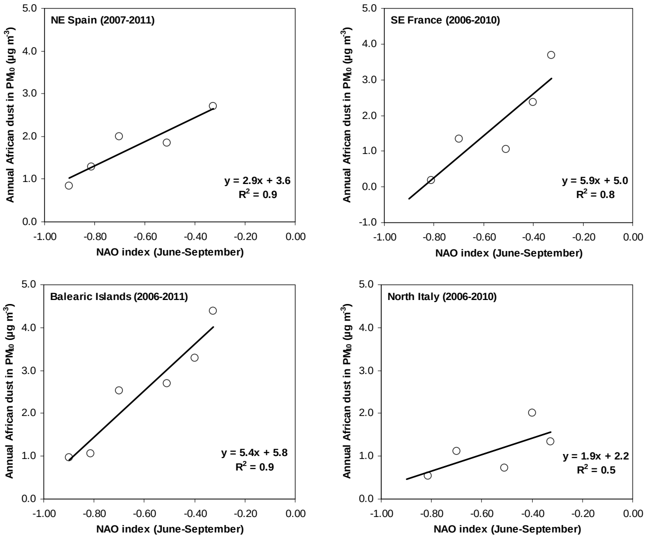

Fig. 12. Scatter-plots between summer NAO index with respect to annual African dust contribution to PM_{10} (in $\mu g\,m^{-3}$) for NE Spain, SE France, the Balearic Islands and north Italy for the period 2006/2007–2010/2011.

contributions are observed, with annual peaks caused by intense episodes. From 2006 or 2007, depending of the area, a marked and continuous decreasing trend is observed in these areas. Because this trend is not observed in the southern areas, a change in the emissions of dust from source regions is not apparent. Nevertheless, a modification in the atmospheric circulation over these areas may be possible. Actually, a recent study on evaluating trends of PM and chemical composition across Europe (Cusack et al., 2012) has demonstrated a partial relation between the observed trends and the North Atlantic Oscillation (NAO) index. The NAO index is accounting for the intensity of the westerly circulation over the north Atlantic (Barnston and Livezey, 1987) that is mainly defined by the position of the two leading atmospheric systems, the Azores high and the Icelandic Low low. Currently, the NAO index is widely used to interpret wintertime weather anomalies in temperature and precipitation over northern, central and western areas across Europe and the United States (Hurrell et al., 2003). In the recent past, such index was evaluated to account for the export of dust (Moulin et al., 1997). In view of the trends found in this work, the NAO index for the summer periods (the dust season in the NW Mediterranean) were calculated from the NOAA data center (http://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/nao.shtml). As seen in Fig. 11a there is a clear change in NAO index from 2006 onwards. In the period 2001–2005, summer NAO indexes were close to 0 whereas there became consistently more negative from 2006–2007 to be at their lowest in 2011 (−0.90). A wider temporal scale of this summer NAO index (Fig. 11b) allows identifying only one persistent negative period in the 50's. Thus, it is evident that the 2006–2011 summer periods were governed by atypical atmospheric patterns. When contrasting the summer NAO index for the period 2006–2011 with respect to the average annual African dust contributions to PM_{10} levels for NE Spain, SE France, the Balearic Islands and north Italy, a perfect fitting is observed (Fig. 12). The correlation observed for these areas is higher in the Spanish areas ($R^2 = 0.9$) than in SE France ($R^2 = 0.8$) or north Italy ($R^2 = 0.5$), thus indicating a progressive lesser influence of the summer NAO index towards the east. Negative NAO phases are related to a displacement of the storm trajectories towards the south. Generally in summer, westerly trade winds blow above 45° in latitude, leaving the Mediterranean region under weak gradient conditions (Millán et al., 1997). During negative NAO phases the track of westerly winds is observed at lower latitude, thus preventing subtropical air masses to reach the NW Mediterranean. In order to address the influence of African dust air masses over summer periods, the geopotential height maps at 850 hPa (Fig. 13a) and the mean temperatures at 850 hPa (Fig. 13b) have been calculated from 2001 to 2011. From 2006 onwards, the Icelandic Low was reinforced and displaced south-easterly (Fig. 13a). Meanwhile, warm air masses (accomplished with mineral dust) emerged from north Africa following atypical trajectories (Fig. 13b). In many cases from 2001 to 2006 such warm air masses attained the western Mediterranean. However, in 2007 and 2008 towards the central Mediterranean occurred, a displacement of such warm air towards the central Mediterranean, although still affecting in 2007 the NE Spain and the Balearic Islands. From 2009 to 2011 north African

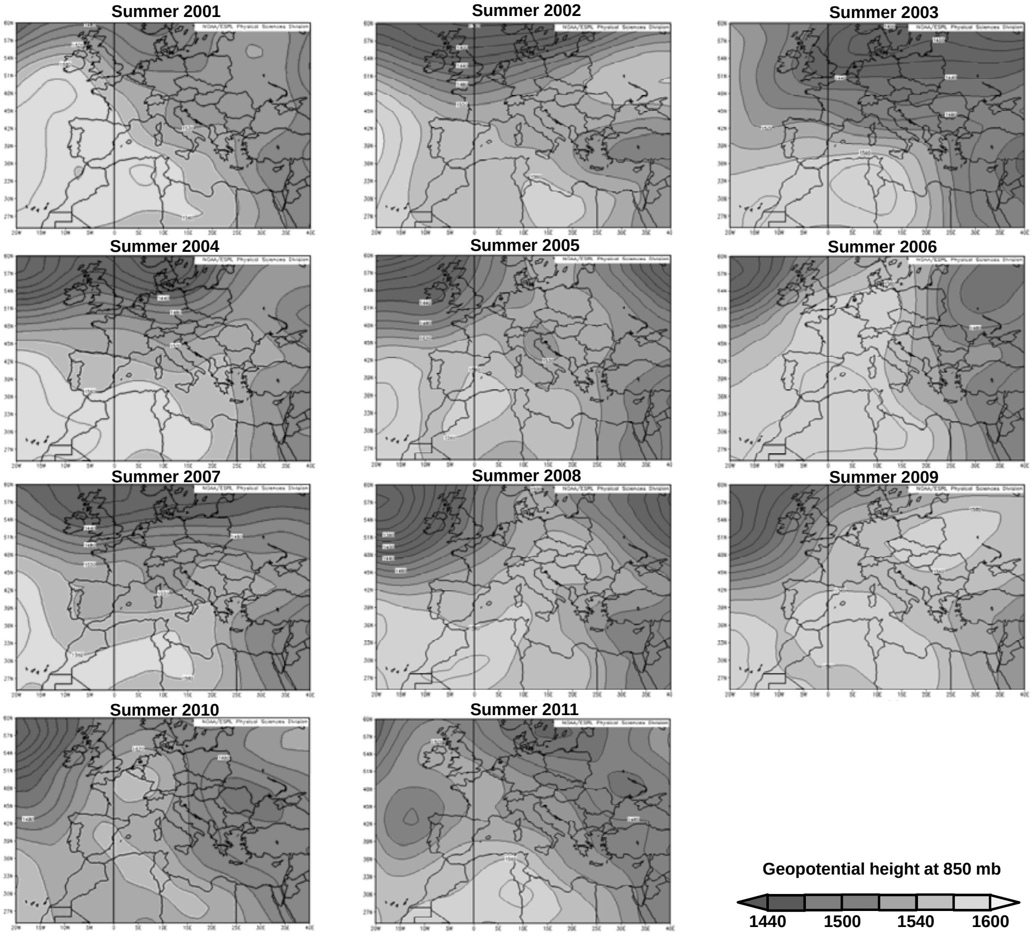
Summer 2001
Summer 2002
Summer 2003
Summer 2004
Summer 2005
Summer 2006
Summer 2007
Summer 2008
Summer 2009
Summer 2010
Summer 2011

Fig. 13a. Mean geopotencial height maps at 850 mb for summer periods from 2001 to 2011.

warm air affected the Iberian Peninsula progressively with lower extent at the same time that it moved towards the NW side of the African continent and the Canary Islands.

4 Conclusions

A comprehensive study on African dust outbreaks occurrence across the Mediterranean Basin and on the influence of such natural source on ambient air PM_{10} levels has been completed. The results of this work reinforce those reported by Querol et al. (2009a) and add new findings that are summarized and discussed in this section:

1. African dust outbreaks across the Mediterranean occur in different seasons and are caused by distinct meteorological scenarios western to eastern of the Mediterranean, which condition their intensity. Intricate and slow-moving transport mechanisms (convective injection of dust in north Africa coupled with anticyclonic conditions at upper atmospheric levels) dominate African dust occurrence, higher in summer, in the western Mediterranean. On the contrary, conventional meteorological scenarios (low pressure systems) provoke a rapid transport of dust towards the eastern Mediterranean, usually from autumn to spring. An intermediate situation occurs in the central part of the Basin, resulting in no significant seasonality but slightly higher number of events in its lower part since is affected marginally by a number of dust outbreaks covering western and eastern parts of the Basin. Accordingly, a number of severe episodes are annually recorded in the eastern part of the Mediterranean, whereas these are scarce or absent in the central and western part.

2. Mean African dust contributions to ambient PM_{10} concentrations across the Mediterranean Basin are varying according to the latitude. A longitudinal effect is patent from 25° E eastwards; with sensibly higher concentrations of African dust in the eastern side of the Basin. Such eastward trend is caused by the occurrence of 2–5 annual severe dust episodes over this area. The Negev

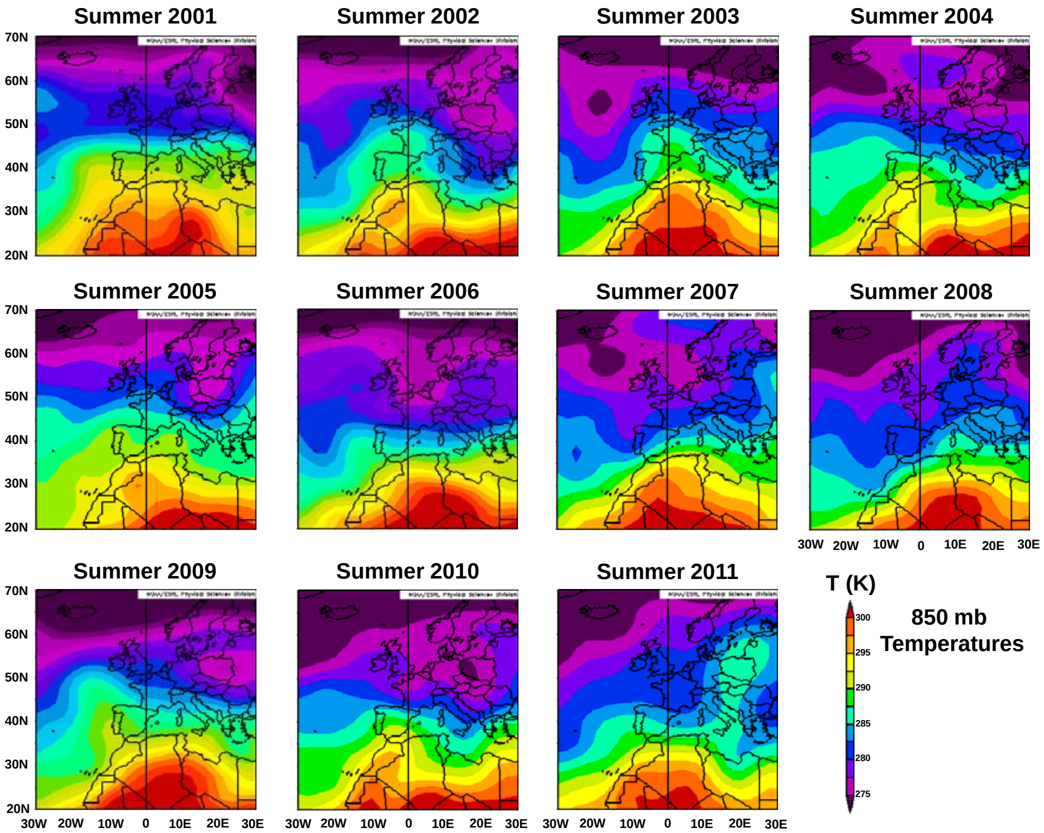

Fig. 13b. Mean temperatures during summer periods (2001–2011), at a geopotencial height of 850 mb.

and the Middle Eastern deserts increase PM_{10} concentrations at Cyprus in about 1 $\mu g\ m^{-3}$.

3. African dust may represent up to 45 % of PM_{10}, as in SE Spain, and around 35 % of PM_{10} in the Greek Islands and Cyprus. Thus, such inputs are considered to be the largest PM_{10} source affecting the regional background of the southern part of the Mediterranean Basin. Over these areas, such natural inputs may represent up to 80 % of PM_{10} during the dust season.

4. Significant inter-annual variations in African dust contributions are observed in the NW of the Mediterranean from 2006/2007 onwards. Such variations are perfectly correlated with the summer NAO index, exceptionally and consistently more negative from 2006 to 2011. Only a similar negative period was observed in the 1950 decade. The averaged geopotential height maps and temperatures calculated at 850hPa for the summer periods from 2001 to 2011 reveal a dislocation of the subtropical warm air (coupled with mineral dust) towards the central Mediterranean in 2007–2008, and towards the NW part of Africa and the Canary Islands in 2009–2011, as well as a strengthening and south-easterly displacement of the Icelandic Low.

Appendix A

It is well known that African dust exerts an important influence on PM concentrations over specific areas (Querol et al., 2009a). In order to compare the RB PM_{10} concentrations avoiding such natural contributions, we have subtracted the African dust estimated in PM_{10} (Fig. A1). Similarly, the high-altitude sites register the lowest PM_{10} concentrations (7–9 $\mu g\ m^{-3}$). Immediately afterwards, western Mediterranean sites show intermediate PM concentrations (10–11 $\mu g\ m^{-3}$). Subsequently, central and eastern Mediterranean locations display higher concentrations (13-15 $\mu g\ m^{-3}$). Accordingly, there is a PM_{10} increase of 3–4 $\mu g\ m^{-3}$ from western towards central and eastern locations across the Mediterranean, parallel to the increase of sulphate and carbonaceous aerosols (Querol et al., 2009b). Finally, the highest PM_{10} concentrations (13–22 $\mu g\ m^{-3}$) correspond to those RB sites located in areas subjected to severe anthropogenic influence (Rome, Marseille and Barcelona).

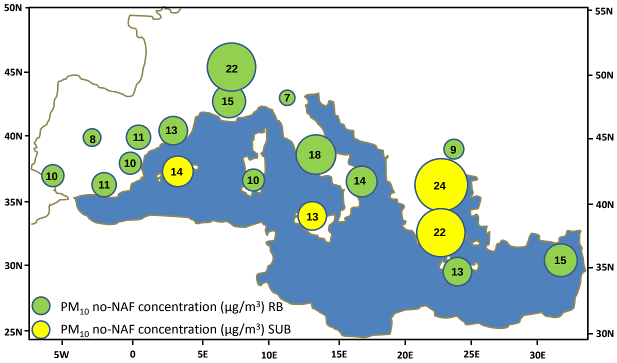

Fig. A1. Annual PM$_{10}$ levels (µg m^{-3}), without African dust, at regional (RB) and suburban (SUB) background sites across the Mediterranean for the period 2001–2011. NAF: African dust outbreaks.

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