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African dust contributions to mean ambient PM_{10} mass-levels across the Mediterranean Basin

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ABSTRACT

Data on mass-levels of PM_{10} measured at regional background sites across the Mediterranean Basin, available from Airbase (European Environmental Agency) and from a few aerosol research sites, are compiled. PM_{10} levels increase from north to south and west to east of the Basin. These variations are roughly coincident with the PM_{10} African mineral dust load. However, when subtracting the African dust from mean PM_{10} levels using a consistent methodology, the PM_{10} background levels are still 5–10 μg m^{-3} higher in the Eastern Basin (EMB) when compared with those in the Western (WMB), mainly due to the higher anthropogenic and sea spray loads.

As regards for the seasonal trends, these are largely driven by the occurrence of African dust events, resulting in a spring-early summer maximum over the EMB, and a clear summer maximum in the WMB, although in this later region the recirculations of aged air masses play an important role. Furthermore, a marked seasonal trend is still evident when subtracting the African dust load. This is characterised by a high summer maximum (driven by low precipitation, high insolation) and a winter minimum (intense synoptic winds).

Important inter-annual variations in the dust contribution are detected, more evident in the southern sites. These differences are generally associated with the occurrence of extreme dust events. Generally, the years with higher dust contributions over the EMB correspond with lower contributions over the WMB, and vice versa.

The characterization of individual particles, collected in both basins during African dust events, by scanning electron microscopy reveals only slight differences between them. This fact probably reflects the high degree of mixture of mineral dust from different sources before the transport towards the receptor sites.

1. Introduction

Crustal aerosols influence the atmospheric radiative balance through scattering and absorption processes (Tegen et al., 1997; Arimoto, 2001; IPCC, 2007), and by acting as cloud condensation nuclei when sulfation and nitration occur (Levin et al., 1996). Dust outbreaks may also greatly increase the ambient air levels of PM recorded in air quality monitoring networks. This is especially relevant in southern Europe (Bergametti et al., 1989; Dayan et al., 1991; Querol et al., 1998; Rodríguez et al., 2001; Escudero et al., 2005, 2007a; Kallos et al., 2006; Mitsakou et al., 2008; Gerasopoulos et al., 2006; Koçak et al., 2007a), Eastern Asia (Zhang and Gao, 2007) and in some Atlantic islands (Prospero and Nees, 1986; Coudé-Gaussen et al., 1987; Chiapello et al., 1995; Arimoto et al., 1997; Viana et al., 2002). Dust particles frequently act as reaction surfaces for reactive gaseous species (Dentener et al., 1996; Levin et al., 1996; Krueger et al., 2004; Alastuey et al., 2005), and the content of secondary PM may greatly increase when dust particles are present in the atmosphere.

Moreover, atmospheric deposition fluxes of specific nutrients in southern Europe are also enhanced by dust outbreaks from Northern Africa (Ganor and Mamane, 1982; Camarero and Catalán, 1993; Rodà et al., 1993; Guerzoni and Chester, 1996; Avila and Rodà, 2002). Oceanic or marine regions may be also highly influenced by crustal dust deposition, when dust iron and phosphate deposition may act as fertilizing agents for phytoplankton (Falkowski et al., 1998; Fung et al., 2000; Arimoto, 2001). Furthermore, chemical

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compounds emitted from deserts may represent a source of alkalinity that neutralizes atmospheric acidity. Finally, dust transport episodes may also cause health impacts due to the high levels of PM and to the transport of anthropogenic pollution (Erel et al., 2006) and also to the possible transport of micro-organisms (Mitsakou et al., 2008; Koulouri et al., 2008; Pérez et al., 2008).

On a global scale, most of the mineral dust is released to the atmosphere from arid or semiarid areas. The major dust source areas are located in subtropical latitudes of the North Hemisphere, and extend from the West coast of North Africa, the Middle East, central and South Asia to China (Prospero et al., 2002).

The low precipitation in the Mediterranean basin favours the long residence time of PM in the atmosphere with the consequent impact on air quality. Furthermore, >70% of the exceedances of the PM_{10} daily limit value (2008/50/CE European directive) in most regional background (RB) EMEP sites of Spain have been attributed to dust outbreaks (Escudero et al., 2007a). Similar findings are mentioned in Gerasopoulos et al. (2006), Koçak et al. (2007a) and Mitsakou et al. (2008) for the Eastern Mediterranean Basin.

According to Escudero et al. (2005), four meteorological scenarios originate the transport of African dusty air masses towards the Western Mediterranean Basin (WMB). These scenarios are characterized by the presence of (1) a North African high located at surface levels (NAH-S), (2) an Atlantic depression (AD) situated in front of Portugal, (3) a North African depression (NAD), and (4) a North African high located at upper levels (NAH-A). During spring and early summer, the development of Saharan thermal lows in the South of Atlas takes place under the influence of the strong thermal contrast between the temperature of the cold marine waters and the warm continental surfaces (Moulin et al., 1998). These cyclones (NAD scenario) travel eastward along this thermal gradient and finally cross the Mediterranean between Libya and Egypt, constituting the main atmospheric scenario responsible for the transport of desert dust over the Eastern Mediterranean Basin (EMB), where also severe episodes can be associated with the combination of a deep trough over West Mediterranean and NW Africa and relatively high pressures to the Eastern part (Kallos et al., 2006).

Escudero et al. (2007b) developed and validated a methodology to determine quantitatively the daily African dust contribution to PM mass-levels recorded in Spain, based on statistical data treatment of PM data series recorded at RB sites. On the other hand, a number of studies on identifying and quantifying dust contributions to ambient PM levels in different parts of the Mediterranean basin are available (e.g. Gerasopoulos et al., 2006; Koçak et al., 2007b; Koulouri et al., 2008; Mitsakou et al., 2008 among others), but as far as we know there are no studies dealing with the entire Mediterranean basin as a whole. Furthermore, differences among local results may be also partially attributable to the different methodologies used.

The aim of the present study is to quantify African dust contributions to mean PM_{10} levels recorded across the Mediterranean basin and to evidence spatial variations. To this end, a common methodology has been applied to PM datasets recorded at two aerosol research monitoring sites (Montseny-Spain and Finokalia-Greece) and at a number of RB sites for a total 21 data series spread across the whole Mediterranean Basin.

2. Methods

Fig. 1 and Table 1 show the location, measurement period and type of air quality monitoring sites from which PM data series have been used in the present study. These are:

a) Aerosol research monitoring sites: Montseny (MSY, NE Spain), and Finokalia (FKL, Crete Island, Southern Greece) EUSAAR (European Supersites for Atmospheric Aerosol Research) sites.
b) Monitoring sites from EMEP (Cooperative Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe) belonging to the air quality monitoring sites of Spain, Cyprus and Bulgaria.
c) Regional air quality monitoring networks, available from Airbase-EEA.

In order to characterize the daily atmospheric scenarios with incidence on PM levels, a number of tools were used:

- NCEP meteorological maps and daily back-trajectories calculated by HYSPLIT4 model (Draxler and Rolph, 2003). Daily 5-days back-trajectories were calculated at 12 h GMT at receptor points of 700, 1500 and 2500 m.a.s.l., by modelling the vertical velocity.
- The occurrence of African dust outbreaks was detected with the previous tools, coupled with the information from the aerosol maps: Marine Meteorology Division of the Naval research Laboratory, USA (NRL) (http://www.nrlmry.navy.mil/aerosol); SKIRON aerosol concentration maps (http://forecast.uoa.gr); BSC-/DREAM dust maps (http://www.bsc.es/projects/earthscience/DREAM/); and satellite imagery provided by NASA SeaWIFS project (http://seawifs.gsfc.nasa.gov/SEAWIFS.html).

Once the PM mass data were obtained, the days under the influence of African dust outbreaks (which will be referred to as NAF) for each receptor site were evidenced with the above methodology. Subsequently, a method based on the statistical data treatment of time series of PM_{10} levels (Escudero et al., 2007b) was used for the quantification of the daily African PM_{10} load during dust outbreaks at each site. The daily RB levels can be obtained by applying a monthly moving 30th percentile to the PM_{10} time series at a RB station after a prior extraction of the days with NAF influence. Then, for a given day under NAF influence, the net dust can be obtained by subtracting the calculated RB value from 30th percentile to the measured PM_{10} concentration. The exclusion of the NAF days for the calculation of the RB levels, and the subsequent estimation of the net dust load may yield an overestimation of the dust load. However, this methodology was validated at three RB sites by comparing the estimated net dust with the experimental crustal load determined in PM_{10} samples (Escudero et al., 2007b). The correlation ($R^2 > 0.86$) and the equivalence (correlation slopes ~ 1) were significant in the three cases.

A statistical analysis was performed on the PM_{10} data collected in the selected stations and mean, standard deviation, relative standard deviation and skewness of the data distributions were derived. Based on a number of samples ranging from about 600 to more than 4000 depending on the station, this kind of statistical analysis allowed for the determination of the mean behaviour of PM_{10} levels and African dust contributions across the Mediterranean basin.

A selection of samples of PM from Spain, Cyprus and Crete, collected during NAF episodes, was also studied under the Scanning Electron Microscope (1450 SEM, JEOL5900LV). Analyses were performed manually on carbon-coated samples using an energy dispersive X-ray microanalysis system (EDX) with a spectrum acquisition time of 30 s live time. Microscope working distance was 10 mm, accelerating voltage 20 kV, and beam current 1 μA.

3. Results

3.1. Temporal and spatial variations

Mean annual PM_{10} mass levels across the Mediterranean revealed clear W–E and N–S increasing trends. Thus, mean PM_{10}

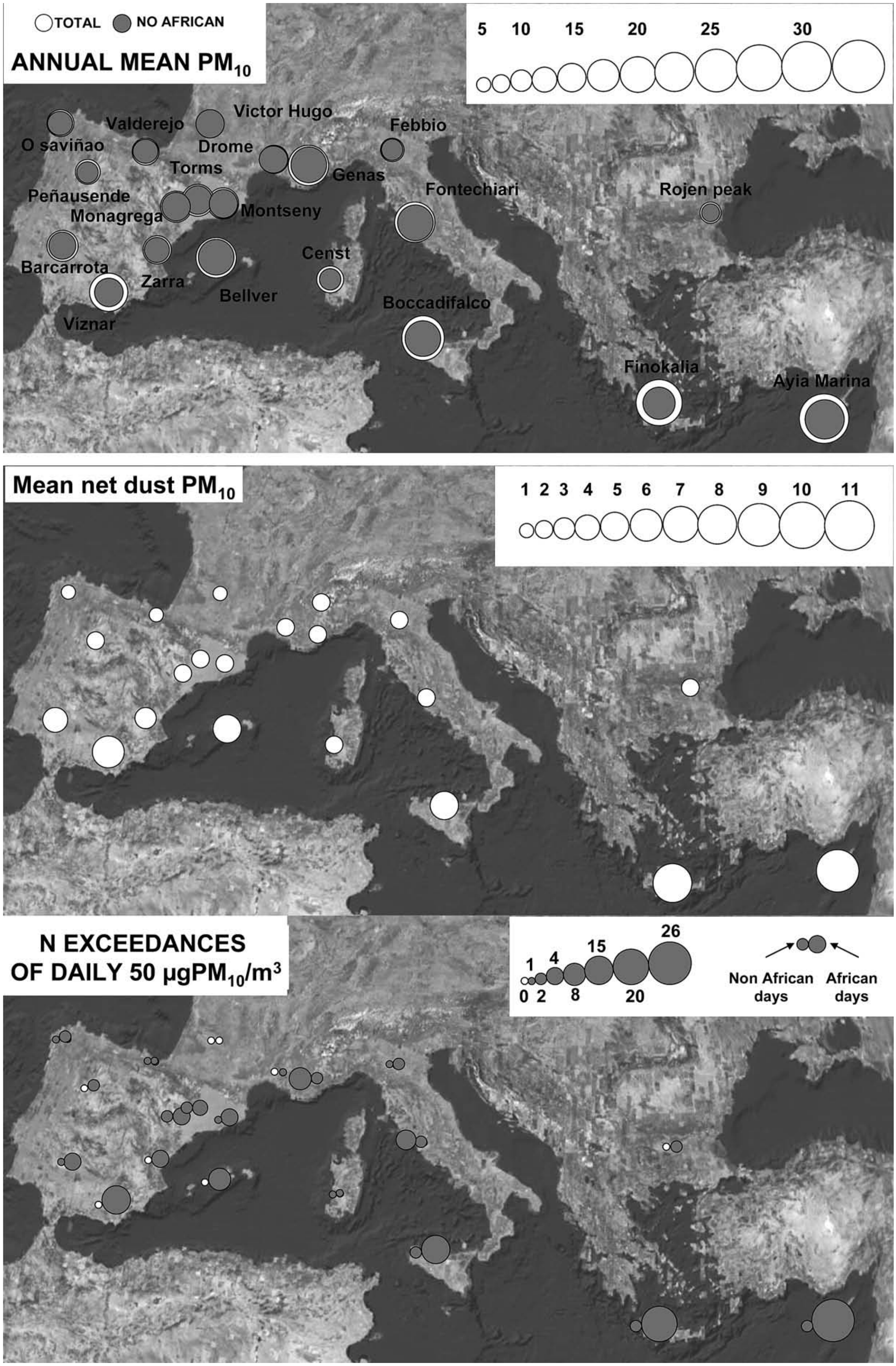

Fig. 1. Location of the PM monitoring sites selected for the study. Top: Mean annual PM_{10} levels for all days (white circles) and excluding the African dust outbreak days (grey circles). Middle: Mean annual African dust contribution to PM_{10} levels. Bottom: Mean annual number of daily exceedances of 50 μg m^{-3} due to African dust (right circle) and due to other causes (left circles).

Table 1
Main characteristics of the selected background stations.

Location	Country	Latitude	Longitude	Altitude (m a.s.l.)	Type of area	Technique	Sampling
Barcarrota	Spain	38° 28′ 33″N	06° 55′ 22″W	393	rural	Gravimetric	Mar 2001–Dec 2007
Viznar		37° 14′ 18″N	03° 28′ 28″W	1265	rural	Gravimetric	Mar 2001–Dec 2007
Zarra		39° 05′ 10″N	01° 06′ 07″W	885	rural	Gravimetric	Mar 2001–Dec 2007
Peñausende		41° 17′ 20″N	05° 52′ 01″W	985	rural	Gravimetric	Mar 2001–Dec 2007
O Saviñao		42° 38′ 05″N	07° 42′ 17″W	506	rural	Gravimetric	Mar 2001–Dec 2007
Valderejo		42° 52′ 31″N	03° 13′ 53″W	911	rural	Beta absorption	Jan 2000–Dec 2007
Bellver		39° 33′ 50″N	02° 37′ 22″E	117	suburban	Beta absorption (Ref. Met. Corrected)	Jul 2001–Dec 2007
Els Torms		41° 23′ 42″N	00° 43′ 16″E	470	rural	Gravimetric	Mar 2001–Dec 2007
Montseny		41° 45′ 36″N	02° 35′ 00″E	728	rural	Laser spectrometer (Ref. Met. Corrected)	Mar 2002–Dec 2007
Monagrega		40° 56′ 48″N	00° 17′ 27″W	570	rural	TEOM	Jan 1996–Dec 2007
Rojen Peak	Bulgaria	41° 41′ 45″N	24° 44′ 19″E	1750	rural	Beta absorption	Jan 2005–Dec 2006
Finokalia	Greece	35° 20′ 00″N	25° 40′ 00″E	150	rural	Beta attenuation	Sep 2004–Dec 2006
Ayia Marina	Cypro	35° 02′ 21″N	33° 03′ 29″E	532	rural	TEOM	Jan 2003–Dec 2006
Victor Hugo	France	45° 15′ 45″N	01° 45′ 46″E	230	rural	Oscillating Microbalance	Jan 2001–Dec 2006
Martigues L'Ile		43° 24′ 18″N	05° 03′ 17″E	1	rural	Oscillating Microbalance	Jan 2001–Dec 2006
Genas		45° 43′ 55″N	04° 58′ 50″E	235	rural	Oscillating Microbalance	Mar 2001–Dec 2006
Drome Rural Sud		44° 31′ 15″N	05° 05′ 24″E	460	rural	Oscillating Microbalance	May 2004–Dec 2006
Fontechiari	Italy	41° 40′ 48″N	13° 40′ 48″E	393	rural	Oscillating Microbalance	Jan 2001–Dec 2006
Boccadifalco		38° 07′ 13″N	13° 18′ 07″E	141	suburban	Beta absorption	Jan 2000–Dec 2006
Sant Antioco (Sardinia)		39° 03′ 52″N	08° 27′ 26″E	270	rural	Beta absorption	Feb 2005–Dec 2006
Febbio		44° 45′ 04″N	10° 26′ 05″E	1020	rural	Beta absorption	Jan 2005–Dec 2006

levels (Table 2 and Fig. 1) range from 15 μgPM_{10} m^{-3} in the W and NW Mediterranean, to near 35 μgPM_{10} m^{-3} in the Eastern basin. An increasing trend is also evident from the Rodope Range and Northern Italy highs (with levels close to 10 μgPM_{10} m^{-3}) to SW, SE and central Mediterranean (22–35 μgPM_{10} m^{-3}).

These W–E and N–S trends are fully coincident with the spatial distribution of the mean annual net African dust contribution to PM_{10}. According to Table 2 and Fig. 1 the mean annual net dust contribution to the annual PM_{10} means recorded at RB sites using the methodology presented at Escudero et al. (2007b), reached mean values of 9–10 μg m^{-3} in the EMB, 6 μg m^{-3} in the SWMB, 2–3 μg m^{-3} in the WMB and <2 μg m^{-3} in the NMB. In addition, the number of days with NAF episodes, with PM_{10} levels exceeding 50 μg m^{-3} (at RB sites), reached mean values of 20–26 days/year in the EMB, 16 days/year in the SWMB, 4 days/year in the WMB and <2 days/year for the NMB. If the days without African dust outbreaks (which will be referred to as no-NAF days) are considered the exceedances are reduced to <3 days/year in most of the Mediterranean. Not only does the frequency of NAF episodes seem to increase easterly, but also in intensity. Thus the mean PM_{10} levels for NAF days reached 51–52 μg m^{-3} in the EMB, 25–36 μg m^{-3} in the S, SW and WMBs, and 19–25 μg m^{-3} in the NMB.

Fig. 2 shows the daily levels of PM_{10} measured at Ayia Marina (Cyprus, representing the EMB) and Monagrega (Eastern Spain, representing the WMB) during 2003–2006, with the indication of NAF days. This figure shows clear seasonal patterns for both the W and EMBs, with markedly higher spring–summer and lower winter PM levels. However, sporadically high PM_{10} levels may be also recorded in winter. A higher frequency and intensity of dust outbreaks is evidenced in the EMB, together with a higher PM_{10} background levels. Thus in the EMBs daily PM_{10} levels during NAF are frequently >100 μg m^{-3} (Fig. 3), and background levels for

Table 2
Mean levels of PM_{10} for the study periods and for the days with (PM_{10} NAF) and without (PM_{10} no-NAF) African dust outbreaks; mean annual number (*N*) of exceedances of the daily PM_{10} limit value (2008/50/CE Directive) for the days with (NAF > 50) and without (no NAF > 50) African dust outbreaks; mean annual net African dust contribution; and mean annual PM_{10} excluding the net dust contribution.

	μg m^{-3}					N of days	
	PM_{10}	PM_{10} no NAF	PM_{10} NAF	net dust	PM_{10}-dust	N no NAF > 50	N NAF > 50
Victor Hugo	16	15	22	1	15	0	0
Martigues L'ile	21	20	27	2	19	2	1
Drome rurale	15	14	25	2	14	0	1
Genas	23	22	28	2	21	7	2
Fontechiari	25	23	32	2	23	6	2
Febbio	11	10	22	2	10	1	2
Rojen peak	10	7	19	2	8	0	2
Sant Antioco	13	11	19	2	11	1	1
Boccadifalco	26	21	38	5	21	2	14
Finokalia	27	19	51	10	17	3	20
Ayia Marina	31	22	52	9	22	2	26
Barcarrota	17	14	28	4	14	1	4
Zarra	15	13	25	3	13	0	4
O Saviñao	14	12	26	2	12	1	2
Valderejo	14	12	24	1	12	1	1
Bellver	24	21	34	3	21	0	8
Els Torms	18	16	27	2	16	2	3
Montseny	18	16	26	2	16	1	4
Monagrega	17	15	30	2	15	2	4
Peñausende	13	10	22	2	10	0	2
Viznar	22	16	36	6	16	0	16

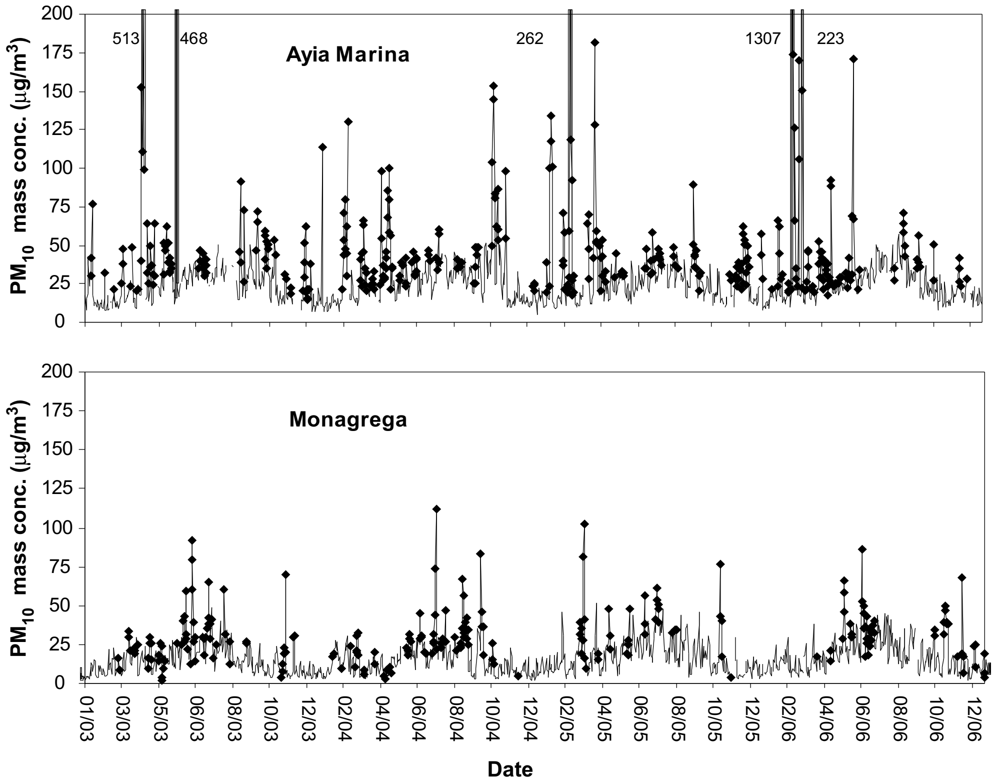

Fig. 2. Daily levels of PM_{10} measured at Ayia Marina (Cyprus) and Monagrega (Eastern Spain) during 2003–2006. Black diamonds indicate days with African dust outbreaks.

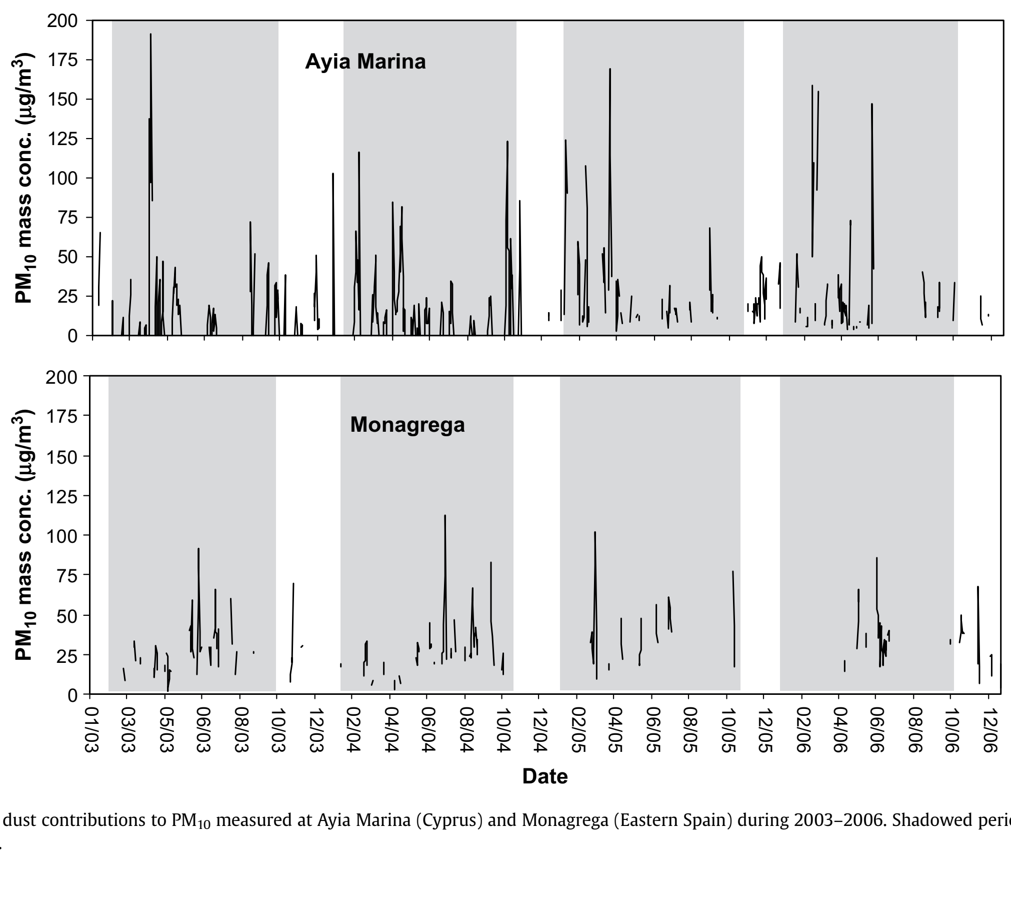

Fig. 3. Daily net African dust contributions to PM_{10} measured at Ayia Marina (Cyprus) and Monagrega (Eastern Spain) during 2003–2006. Shadowed periods indicate February to October (both included).

winter and summer no-NAF reached 15 and 35 μg m^{-3} (Fig. 4), respectively. However, in the WMB, daily PM_{10} levels during NAF are frequently <100 μg m^{-3}, and background levels for winter and summer lower than 10 and 25 μg m^{-3} (Figs. 3 and 4). The observed seasonality and spatial variability of African dust contributions extended at the whole stations will be discussed in the following sections.

The differences observed in PM_{10} background in both warm and cold seasons should be attributed to the higher annual sulphate and sea spray levels recorded in the EMB, compared to the WMB: around 5.5 and 2.5 μg m^{-3} for sulphate, 3–8 and 1–2 μg m^{-3} for sea spray respectively, for the annual means at the E and WMB, according to Querol et al. (2009). Furthermore, RB levels of carbonaceous particulate matter were also slightly elevated in the EMB with respect to the WMB (Querol et al., 2009). Thus, the results show that the background levels of anthropogenic pollutants are also higher in the EMB compared to WMB. This may be due to emissions from coal power generation in Greece and Turkey, but also to the high influence of air masses coming from Eastern Europe (Ukraine, Russia) (Kallos et al., 1993, 1998; Mihalopoulos et al., 1997; Gerasopoulos et al., 2005; Lelieveld et al., 2002; Erel et al., 2007). Sea spray levels may be higher in the EMB due to the strong northerly winds "Etesians" induced by the Indian Monsoon (Kallos et al., 1993; Rodwell and Hoskins, 2001) and the complex orographic terrain of regions such as Greece. Conversely, during summer, in the WMB atmospheric stagnation and recirculation episodes are developed in the WMB, simultaneously with the Etesian dominated scenario in the EMB (Millan et al., 1997; Kallos et al., 2007).

As regards the seasonality of African dust outbreaks in the EMB, Figs. 2 and 3 show clearly that the lowest frequency of African dust outbreaks occur from November to January; and that the maximum take place from late winter to spring, with a slightly lower frequency in summer. A similar pattern was observed for the WMB, but with summer episodes generally higher in frequency and intensity.

The spring–summer PM maxima are associated with the higher frequency of African dust outbreaks, lower precipitation, higher (local/regional) resuspension owing to the dryness of soils, increased formation of secondary aerosols caused by the maximum solar radiation, and recirculation of air masses that prevent air renovation (in the WMB). The sporadic second-order winter maximum in the WMB is attributed to intense pollution anticyclonic episodes of anthropogenic PM and sporadic winter NAF episodes. The seasonal variation of PM levels at RB sites is also influenced by the evolution of the boundary layer. Thus, at Montseny, when the monitoring site is outside the boundary layer (especially in winter), it is less affected by regional anthropogenic emissions. In contrast, the high vertical development of the boundary layer in summer allows PM pollutants to reach the monitoring site.

If the NAF days are excluded from the calculation of the mean PM_{10} annual levels (Table 2 and Fig. 1), the spatial variations across the Mediterranean are much reduced. Thus, the highest levels (19–23 μg m^{-3}) were now recorded in areas: a) where local anthropogenic influence is evidenced (regional-suburban background sites such as Genas, Fontechiari, Boccadifalco and Bellver), or b) in island or coastal sites (due to the sea spray contribution, Ayia Marina, Bellver, and Finokalia). At most of the other RB sites, mean PM_{10} levels ranged from 11 to 16 μg m^{-3}, with the exception of the high mountain areas of the NMB (Rojen Peak) where levels <10 μg m^{-3} are usually measured.

PM_{10} speciation data compiled by Querol et al. (2009) for Montseny and Finokalia, and by Rodriguez et al. (2004) for

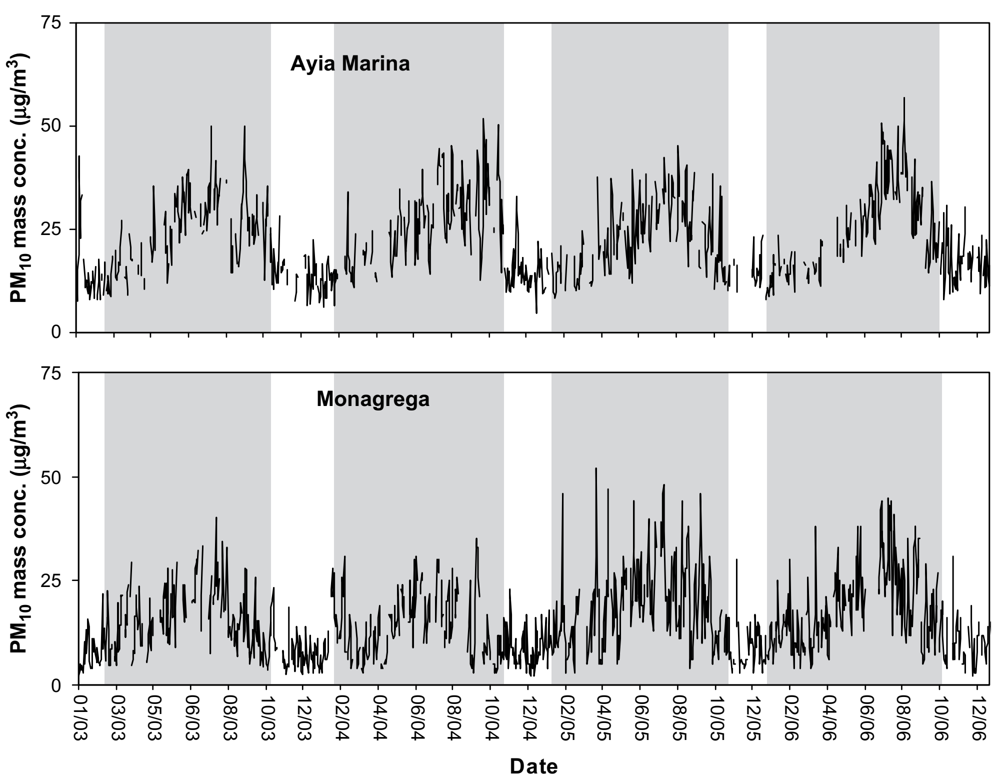

Fig. 4. Daily levels of PM_{10} measured at Ayia Marina (Cyprus) and Monagrega (Eastern Spain) during days without African dust outbreaks in 2003–2006. Shadowed periods indicate February to October (both included).

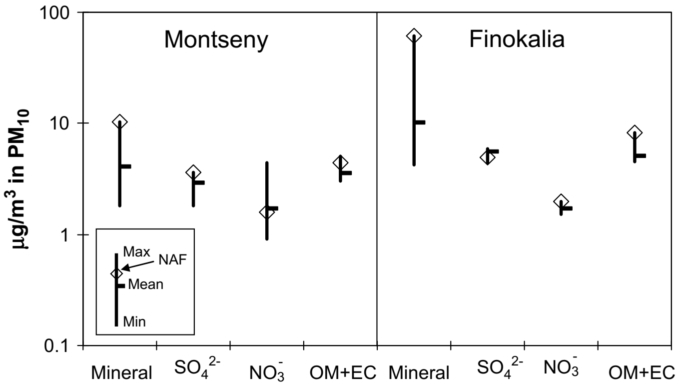

Fig. 5. Maximum, minimum, mean levels of mineral matter, sulphate, nitrate and carbonaceous aerosols measured at Montseny and Finokalia for the different atmospheric transport scenarios according with data on PM speciation from Querol et al. (2009). Diamonds indicate the mean levels recorded at both sites for the African dust transport scenario.

Monagrega show that the mean annual contribution for total mineral matter reached 4, 14 and 4 µg m^{-3} for these 3 sites, respectively. On the other hand, in the present study we deduced that the annual net dust contribution reaches 2 (2002–2007), 10 (2004–2006) and 2 (1996–2007) µg m^{-3}. Based on these values we might suggest that there is a RB mineral matter of around 2 and 4 µg m^{-3} in the W and EMBs, respectively, that it is not attributable to African dust transport, but rather to the regional- (and local-) scale resuspension of dust.

As shown in Fig. 5 the highest mineral levels at Montseny and Finokalia were measured during NAF episodes (14 and 60 µg m^{-3}). Also the carbonaceous aerosols showed relatively high levels during the African episodes, probably due to the poor dispersive conditions and to the mixture of dust with other anthropogenic emissions from Northern Africa (including possible biomass burning). At Montseny, the highest sulphate levels were also recorded during African dust and regional recirculation episodes due to the sulfation of dust (Levin et al., 1996). Conversely, the lowest levels of mineral matter, sulphate, nitrate and carbonaceous material were measured during Atlantic advection episodes (Querol et al., 2009).

3.1.1. Statistical analysis

For the PM_{10} mass data collected in the study stations (Table 1) a statistical approach was used in order to determine the mean aerosol characteristics at each site. In Table 3 the number of samples, mean PM mass levels, standard deviation, relative standard deviation and skewness of the calculated data distributions were reported for the whole periods and separately for the summer (April–September) and winter (October–March) periods. Relative standard deviations in the order of 40–80% were observed at all stations. The highest fluctuations were observed for Ayia Marina (145%) and Finokalia (148%) mainly because of the exceptionally high PM levels registered in these stations as a consequence of a higher occurrence of severe Saharan dust intrusions in the EMB. On average, as already stated, the PM_{10} levels showed seasonal variations with higher values in summer than in winter. Winter PM levels of 30–50% lower than the summer ones were observed for Peñausende, Valderejo, Barcarrota, Zarra, Monagrega, Viznar and Rojen Peak. Variations of about 10–30% were registered at Els Torms, Drome rurale, Bellver, Ayia Marina, Febbio, Boccadifalco, O Saviñao, Sant Antioco and Montseny. An opposite condition was instead observed in Fontechiari,

Table 3
Characteristic quantities of PM_{10} levels in the considered stations.

Station	Whole period (Table 1)					Summer				Winter			
	N	mean	*σ*	relative *σ*	sk	*N*	mean	*σ*	relative *σ*	*N*	mean	*σ*	relative *σ*
Barcarrota	2352	17.3	13.1	0.76	3.7	1226	20.9	13.5	0.65	1126	13.5	11.5	0.85
Viznar	2375	21.9	17.1	0.78	3.2	1239	27.7	17.8	0.64	1133	15.5	13.0	0.84
Zarra	2393	15.4	10.3	0.67	2.8	1237	18.6	10.0	0.54	1156	12.0	9.6	0.80
Peñausende	2346	12.5	10.6	0.85	3.5	1230	14.7	11.0	0.75	1116	10.0	9.6	0.96
O Saviñao	2293	13.7	9.6	0.70	2.3	1218	15.4	9.8	0.64	1075	11.8	9.0	0.76
Valderejo	2882	13.6	8.1	0.60	2.3	1454	16.2	8.4	0.52	1428	10.9	6.9	0.63
Bellver	2199	23.6	12.6	0.53	3.3	1100	26.0	11.5	0.44	1099	21.2	13.3	0.63
Els Torms	2315	18.2	10.6	0.58	1.8	1203	19.2	9.6	0.50	1112	17.0	11.5	0.68
Montseny	1871	17.5	10.4	0.60	1.1	955	19.9	9.8	0.49	916	14.7	10.3	0.70
Monagrega	4330	17.0	11.8	0.69	1.7	2175	21.2	12.3	0.58	2155	12.8	9.6	0.75
Rojen Peak	583	9.5	8.5	0.89	1.6	314	12.6	9.0	0.71	269	5.8	6.0	1.03
Finokalia	603	27.5	40.8	**1.48**	5.5	284	27.6	32.3	1.17	319	27.4	47.1	1.72
Ayia Marina	1378	30.7	44.6	**1.45**	6.5	689	34.1	30.4	0.89	689	27.4	55.2	2.01
Victor Hugo	2122	16.4	6.2	0.38	1.0	1088	15.7	5.4	0.34	1034	17.2	6.9	0.40
Martigues	1658	20.7	9.3	0.45	1.3	875	20.4	7.9	0.39	782	20.9	10.7	0.51
Genas	1873	22.6	11.4	0.51	1.6	971	20.2	8.2	0.41	902	24.8	13.6	0.55
Drome rurale	865	15.3	8.2	0.54	1.2	456	16.5	8.2	0.50	409	14.0	7.9	0.56
Fontechiari	1342	24.5	11.6	0.47	1.8	650	21.3	8.4	0.39	692	27.4	13.3	0.49
Boccadifalco	1767	25.5	16.4	0.64	3.5	838	28.8	13.2	0.46	929	22.4	18.2	0.81
Sant Antioco	648	12.6	7.7	0.61	1.8	358	14.3	7.9	0.55	290	10.6	6.9	0.65
Febbio	677	11.4	9.1	0.80	1.4	333	12.7	9.2	0.72	344	10.1	8.9	0.88

Genas, Victor Hugo and Martigues with winter mean PM_{10} values respectively of 30%, 23%, 10% and 3% higher than those measured in summer. This behaviour could be attributed mostly to local conditions with possible important pollution episodes increasing the PM_{10} levels during the coldest months. It can be noted from Table 3 that the relative standard deviations were always higher in winter than in summer at all stations, thus evidencing relative higher fluctuations in the PM_{10} levels during the coldest months in the entire Mediterranean basin.

All the calculated distribution functions of the PM_{10} levels show positive skewness (Table 3) as typical for many positive-defined meteorological parameters (O'Neill et al., 2000; Matthias and Bösenberg, 2002; Behnert et al., 2007). The skewness measures the asymmetry of a distribution function and the higher the skewness, the higher the probability of measuring high PM levels in the corresponding measurement site. High skewness was observed especially in Ayia Marina (6.5) and Finokalia (5.5) as a consequence of high occurrence of extreme NAF episodes with contributions to the PM mass much higher than the calculated PM_{10} mean. In Ayia Marina and Finokalia PM_{10} levels higher than 700 $\mu g\ m^{-3}$ and 1300 $\mu g\ m^{-3}$ were respectively observed and the number of daily PM_{10} level higher than 100 $\mu g\ m^{-3}$ was the highest registered in the Mediterranean basin. Skewness between 2 and 4 was observed for Barcarrotta, Viznar, Zarra, Bellver, Peñausende and Boccadifalco indicating the relatively high occurrence of high PM_{10} levels in the entire SMB. For the aforementioned stations the skewness calculated on the no-NAF data distributions strongly reduced at values ranging from about 0.5 (Ayia Marina, Zarra and Viznar) to about 2 (Boccadifalco and Barcarrota) thus demonstrating the high influence of the Saharan dust episodes. In the NMB the skewness was considerably lower than 2, except for O Saviñao and Valderejo (Table 3), with no-NAF skewness only slightly lower thus indicating a reduced effect of the dust pollution episodes in the NMB. Skewness values of 2.3 in O Saviñao and Valderejo indicated some degree of pollution episodes different from the desert dust ones. Consequently, as shown in Fig. 6, a general increasing trend was observed for the calculated skewness as a function of locations within the Mediterranean Basin and strength of dust pollution episodes (net dust).

3.2. Inter-annual trends in dust contribution

Important inter-annual differences in the dust contribution were registered in the WMB and EMB. Thus, considering the short study period 2004–2006, the mean annual net dust contribution at the RB site of Ayia Marina (EMB) ranges from 8 to 12 $\mu g\ m^{-3}$, and from 6 to 9 $\mu g\ m^{-3}$ at Viznar (WMB, Fig. 7). Note also that the maximum monthly net dust contributions recorded in the WMB are usually coincident with the lowest registered in the EMB, and vice versa (Fig. 7). The detailed study of the meteorological phenomenology of this period (2004–2006) allows us to attribute these differences to the frequency of occurrence of the specific meteorological scenarios responsible for the transport of African dusty air masses, but also to the intensity of some episodes, especially in the EMB.

Fig. 7 shows the mean monthly dust contribution at Ayia Marina (EMB) and Viznar (WMB) from 2004 to 2006. Simultaneously, the number of days with African dust influence is also represented. Firstly, the seasonal distribution of the African dust events is different in the two ends of the basin. The EMB registers the highest frequency of occurrence in winter and spring, whereas in the WMB this occurs in summer. This shows that the dust transport towards the WMB and EMB is associated with synoptic conditions that are in opposite phase. Second, there is a positive relationship between the number of African dust outbreaks and the net dust registered (Fig. 7) Nevertheless, some intense episodes may alter this correlation. Third, although a seasonal distribution of the dust contribution may be deduced from the results, an important variability has been identified along the different years studied.

Fig. 8 shows some meteorological scenarios responsible for the transport of African dusty air masses towards the WMB and the EMB. Fig. 8a shows the mean sea level pressure at surface from 1st to 31st March 2004. The low pressures over North Africa forced the dusty air masses towards the whole Mediterranean basin. The net dust (in PM_{10}) calculated for this month reaches around 13 $\mu g\ m^{-3}$ at both W and EMB sites. Fig. 8b illustrates the mean sea level pressure at surface from 1st to 31st March 2006. In this case the EMB was affected by a North African depression, responsible for the transport of dusty air masses, whereas the

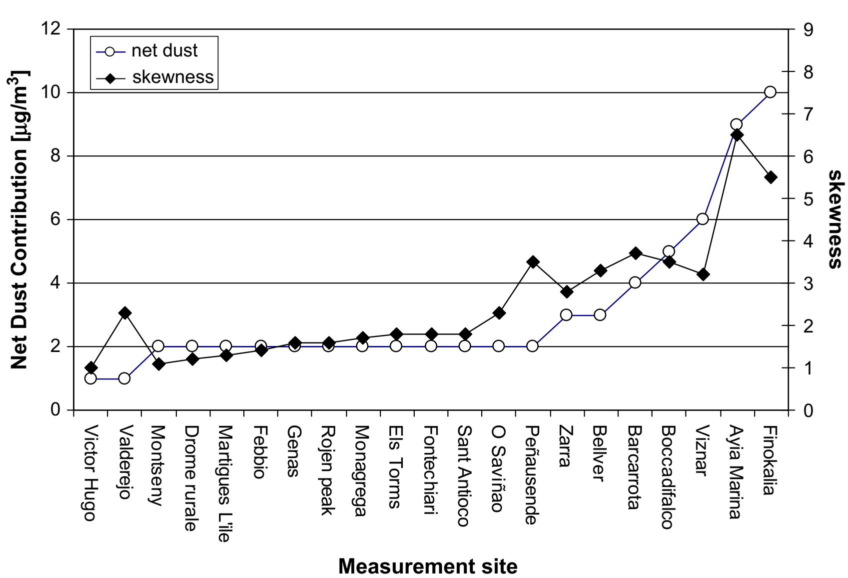

Fig. 6. Calculated skewness and net dust contributions for each station.

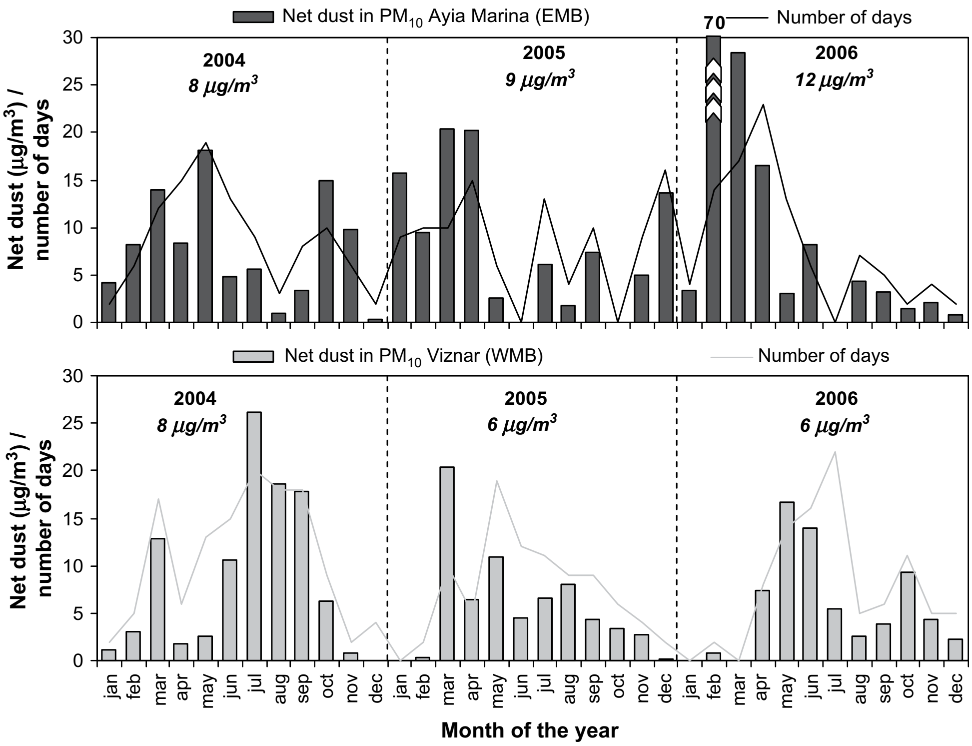

Fig. 7. Mean monthly net dust in PM_{10} (shadowed bars) from 2004 to 2006 calculated for Ayia Marina-Cyprus (EMB) and Víznar-Spain (WMB). Number of days with African dust outbreaks influence (line). Mean annual net dust contributions are shown for each year.

WMB was under the influence of Atlantic winds as a result of the proximity of a high pressure system to the coast of Portugal. During this period the EMB registered 15 days with African dust influence and a mean net dust contribution of 28 µg m^{-3}, whereas no African dust episodes were recorded in the WMB. Fig. 8c illustrates the typical summer scenario (August 2004) showing a high pressure system over Tunisia at 850 mb of geopotential height that favours the transport of African air masses towards the WMB, whereas winds from central Europe blow over the EMB. During August 2004 the EMB registered only 4 days of African dust air masses and a mean contribution of 2 µg m^{-3}, whereas the WMB recorded 17 African dust events and a mean contribution of 18 µg m^{-3}. Finally, Fig. 8d shows a summer scenario occurred in August 2006. In this case the high pressure system typical of Tunisia was absent. The whole Mediterranean was affected by western and northern winds, and consequently the African dust was scarce. Around 4–6 events were registered in all cases, with a mean contribution of 3–5 µg m^{-3}.

3.3. African dust single particle composition: SEM studies

Samples from west and east Mediterranean were studied under the SEM in order to compare the chemical composition of individual particles during the intrusion of air masses from North Africa. Following the approach and methodology explained in Moreno et al. (2003) where SEM limitations are discussed one can compare the relative importance of mineralogical groupings between samples, based on elemental ratio values, differentiating between felsic silicates (Si- and Al-rich, such as quartz, feldspars, white micas or clays) from more mafic silicates (rich in Fe and Mg, as serpentine, talc, vermiculite or amphiboles). The samples selected were PM_{10} and $PM_{2.5}$ filters from Cyprus (Larnaka, Pafos, Strovolos), Crete (Finokalia) and NE Spain (Montseny).

A total of 600 particles (PM_{10} and $PM_{2.5}$) were identified from samples collected at each of the 3 sites in Cyprus (November 2008), with no significant differences between the overall major element chemistry of particle populations analysed at the 3 sites. Silicates are dominant (50–55% of PM_{10}, 70–75% of $PM_{2.5}$), followed by carbonates (33–40% PM_{10}; 15–22% $PM_{2.5}$), sulphates (up to 13%) and sea salt (up to 8%) (Fig. 9a). With regard to silicate composition, Al-rich clay minerals such as kaolinite and illite (Al-felsic silicates on Fig. 9b) are very common in the PM_{10} silicates, whereas, siliceous compositions likely to represent quartz or biological silica (e.g. diatoms) are more abundant in $PM_{2.5}$. Compositions in between these two groups (Si felsic on Fig. 9b), representing feldspathic compositions, Si-rich clays, and mixed analyses, form 35–46% of all silicates with no marked difference between coarse and fine samples.

A total of 800 particles from the Crete PM_{10} samples (April and November–December 2008) were similarly identified. Most of these particles are felsic silicates (clay minerals, silica and feldspar: 74%), with lesser amounts of carbonates (up to 15%), sulphates (up to 11%), and mafic silicates (4%). Al-rich clays compositions (such as kaolinite and illite) are clearly dominant over Si felsic and silica compositions (Fig. 9b). We can conclude overall that the Cyprus PM_{10} samples are more siliceous and less Al-clay rich than those collected from Crete. Sea salt was detected as a common component but only in some of the samples (up to 9%). There are no obvious differences between those samples collected during spring (April) and winter (Nov–Dec) dust intrusions.

A total of 600 individual mineral particle analyses were obtained from NE Spain (Montseny) samples collected during NAF episodes from Central Africa (July 2004) and NW Africa (February 2004). The PM_{10} fraction of these samples is dominated by silicate PM_{10} (92–

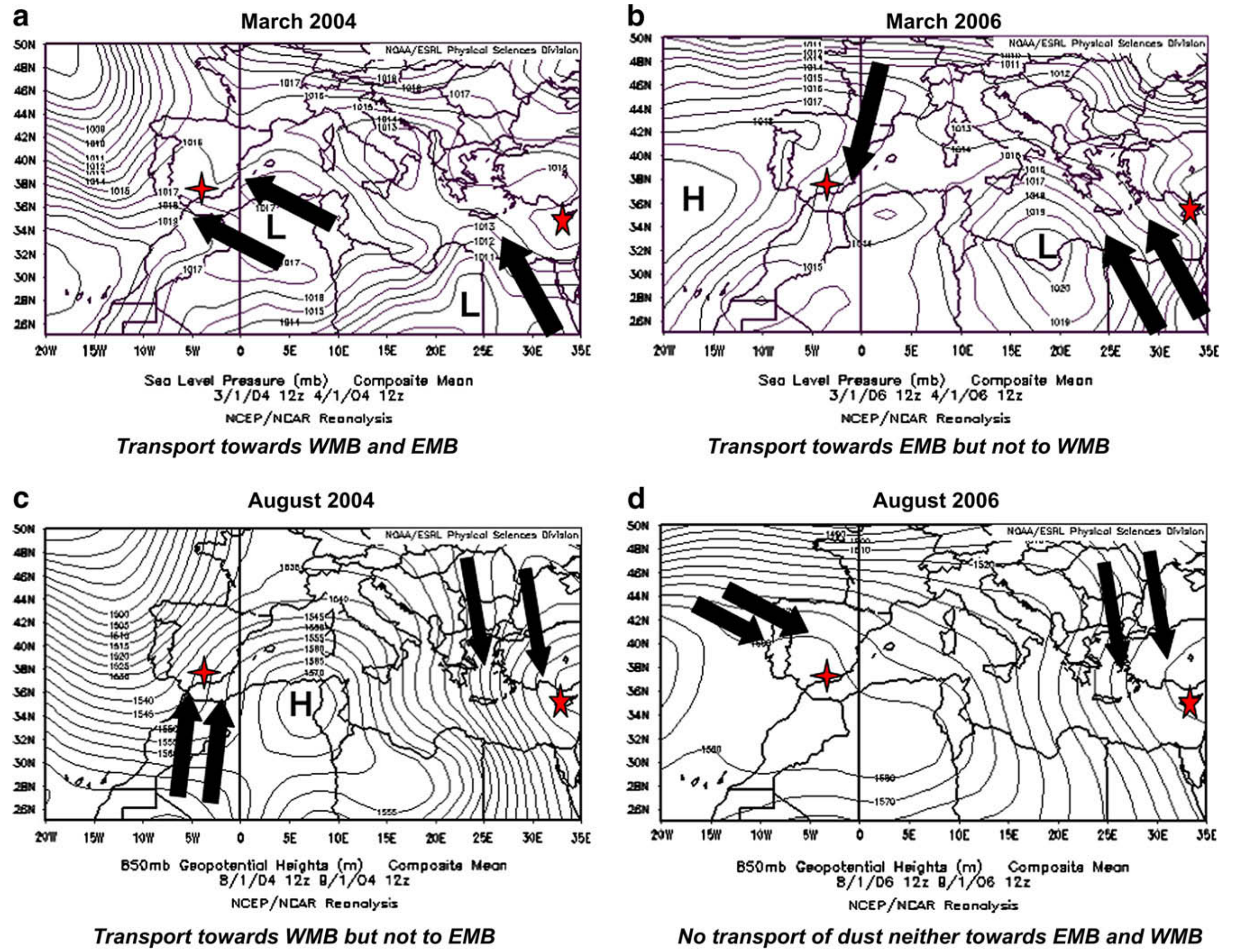

Fig. 8. Examples of some meteorological scenarios favouring or preventing the transport of African dusty air masses towards the WMB and the EMB. The four-peak star locates Víznar (Spain) and the five-peak star situates Ayia Marina (Cyprus).

97% in number), with low proportions of Ca- (and Mg-) carbonates (1–4%), sulphates (1%) and carbonaceous compounds (1–3%) (Fig. 9a). These proportions are different in the $PM_{2.5}$ fraction which has much fewer silicate particles (up to 67%), and many more carbonaceous PM (up to 26%), whereas sulphate content is similar between the two PM fractions. Aluminous felsic silicates, presumably mostly kaolinitic and illitic-smectitic clays (Kalderon-Asael et al., 2009; Moreno et al., 2006) are more common in the samples sourced from Central Africa than that from NW Africa.

Thus, SEM results show Cyprus as the most calcareous airborne samples (presumably they are richest in calcite), with silicates being more common in the $PM_{2.5}$ fraction and carbonate in the coarser fraction (PM_{10}), and with no significant compositional differences between the 3 collecting sites during the same intrusion episode. Crete samples on the other hand are the richest in Al-clays (e.g. kaolinite and illite) and show no obvious differences between samples collected during NAF events occurred in different times of the year (April vs. November–December). The latter observation is perhaps not surprising as the back-trajectories for the different episodes only indicate slight differences in the source areas. Finally the samples from Montseny (NE Spain) are the least calcareous (<5% carbonate), and in this sense are chemically more similar to the Crete samples rather than the Cyprus samples. With regard to silicate composition, the closest similarities are between the airborne samples collected on Crete and those arriving in NE Spain from Central North Africa (Fig. 9b).

4. Conclusions

Regional background PM_{10} levels across the Mediterranean show clear increasing trends from north to south and west to east of the Basin. These trends are almost coincident with the PM_{10} African dust load. Independently, when subtracting the PM load attributed to the African dust, the background levels are 5–10 $\mu g\ m^{-3}$ higher in the EMB when compared with those in the WMB, mainly due to the higher background contributions of anthropogenic compounds (sulphate and organics) and sea spray.

Marked PM_{10} seasonal trends are evidenced, largely driven by the occurrence of African dust events. Thus, the higher frequency of dust events in spring–early summer over the EMB causes higher PM levels. Likewise, the summer maximum observed in the WMB roughly coincides with the most intense period of African dust outbreaks, but in this region also with the development of recirculations of aged air masses. Furthermore, a marked seasonal trend is still evident when subtracting the African dust load. This is characterised by a high RB summer maximum (driven by low precipitation, high insolation) and a winter minimum.

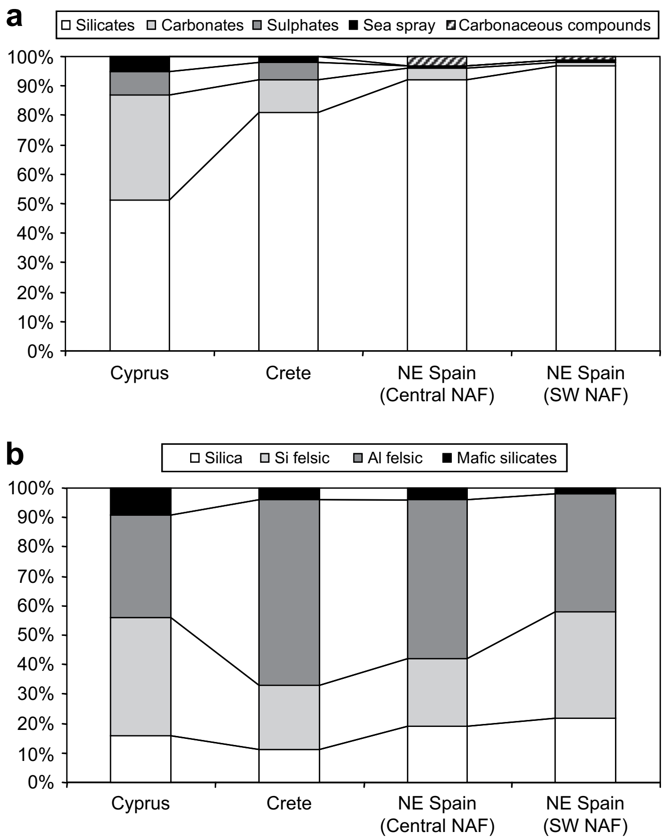

Fig. 9. a) SEM-EDX PM composition (in % number of particles, not mass) for PM_{10} filter samples collected during north African pollution episodes in locations from Cyprus, Crete and NE Spain (the latter including air masses coming from central and SW Africa). b) Idem, considering only silicate compositions (depending on the Al/Si and (Mg + Fe + Al + Si)/Si values, species are inferred in % number of particles).

This study also points out important inter-annual variations in the dust contribution, more evident in the southern sites. These variations are generally associated with the occurrence of extreme dust events. Generally, the years with unusually high dust contributions over the EMB corresponds with anomalously low contributions over the WMB, and vice versa.

The statistical data treatment reveals a higher probability of elevated PM levels (>100 $\mu g\ m^{-3}$) occurrence in the EMB rather than in the WMB owing to the higher occurrence and intensity of African dust intrusions. A positive correlation is detected between elevated skewness values and occurrence of extreme PM events, always associated to the impact of African dusty air masses.

Finally, the characterization of individual particles, collected during African dust events, by scanning electron microscopy reveals only slight differences between them. This fact probably reflects the high degree of mixture of mineral dust from different sources before the transport towards the receptor sites.

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