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PM_{10} and $PM_{2.5}$ Levels in the Eastern Mediterranean (Akrotiri Research Station, Crete, Greece)

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Abstract Particulate matter measurements (PM_{10}, $PM_{2.5}$) using a beta radiation attenuation monitor were performed at the Akrotiri research station (May 2003–March 2006) on the island of Crete (Greece). The mean PM_{10} concentration during the measuring period (05/02/03–03/09/04) was equal to 35.0±17.7 μg/m^3 whereas the mean $PM_{2.5}$ concentration (03/10/04–04/02/06) was equal to 25.4±16.5 μg/m^3. The aerosol concentration at the Akrotiri station shows a large variability during the year. Mean concentrations of particulate matter undergo a seasonal change characterised by higher concentrations during summer [PM_{10}, 38.7±10.8 μg/m^3 (2003); $PM_{2.5}$, 27.9±8.7 μg/m^3 (2004) and 27.8±9.7 μg/m^3 (2005)] and lower concentrations during winter [PM_{10}, 28.7±22.5 μg/m^3 (2003/2004); $PM_{2.5}$, 21.0±13.0 μg/m^3 (2004/2005) and 21.4±21.9 μg/m^3 (2005/2006)]. Comparative measurements of the PM_{10} concentration between the beta radiation attenuation monitor, a standardized low volume gravimetric reference sampler and a low volume sequential particulate sampler showed that PM_{10} concentrations measured by the beta radiation attenuation monitor were higher than values given by the gravimetric samplers (mean ratio 1.17±0.11 and 1.21±0.08, respectively). Statistical and back trajectory analysis showed that elevated PM concentrations (PM_{10}, 93.8±49.1 μg/m^3; $PM_{2.5}$: 102.9±59.9 μg/m^3) are associated to desert dust events. In addition regional transport contributes significantly to the aerosol concentration levels whereas low aerosol concentrations were observed during storm episodes.

Keywords Aerosol measurements · Back trajectory analysis · Crete · PM_{10} · $PM_{2.5}$

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1 Introduction

Long-range transboundary transport is responsible for a significant fraction of the particulate pollution in European cities (WHO/UN 1999; EC 2004; EMEP

2003, 2006). A major part of this contribution is secondary particulate matter (PM) in the form of sulphate, nitrate, ammonium and organic aerosols formed by the oxidation of sulphur dioxide, nitrogen oxides and organic gaseous species. Recent scientific efforts are focused on the quantification of particulate matter emissions, the long-range transport of primary particulate matter and the formation of secondary organic particulate matter from natural and anthropogenic emissions (EC 2004; EMEP 2006). Especially, on the PM phenomenology in Europe two studies have been published (Van Dingenen et al. 2004; Putaud et al. 2004). However, an aspect that is missing in the above studies is the quantification of particulate matter emissions from natural sources, such as sea-salt aerosol emissions and Aeolian dust transport from the Sahara in Southern Europe.

As published earlier (EMEP 2006), there is a consistent pattern in the geographical variability of PM concentrations in Europe consisting of comparable lower concentrations of particulate matter in the far north, high concentrations in central Europe and of higher concentrations in southern and central countries. This is due to elevated anthropogenic emissions in central Europe as well as due to differences in the weather conditions (e.g. rainfalls, solar radiation) and the vegetation in southern countries. Moreover, this variability results from continuing high emissions of anthropogenic gaseous and aerosol pollutants in Southern Europe and of very reactive unsaturated hydrocarbons (including isoprene) of natural emissions (Hoffman et al. 1997). In addition, in our study area, the Mediterranean, sea spray and North African desert dust contribute significantly to the aerosol load at specific time periods: sea-salt during winter and summer time and dust during spring and autumn. These natural particulate emissions are involved in heterogeneous reactions with anthropogenic gaseous pollutants and can modify the processes leading to gas-to-particle conversion (Millan et al. 1997; Rodriguez et al. 2002; Bardouki et al. 2003).

At the north of the Mediterranean Sea a number of highly populated European countries exist with industrial, semi-industrial, and rural economies, whilst to the south is Africa. A detailed wind trajectory analysis shows that more than 60% of air masses, which arrive in the Eastern Mediterranean, come from the north–northwest (N–NW) and 13–16% from the Sahara (Guerzoni et al. 1990; Mihalopoulos et al. 1997). Air masses from the N–NW contain particles related to industrial and urban inputs; those from Sahara carry predominantly mineral dust. Transport of desert dust occurs mostly during the spring and summer and causes non-continuous crustal aerosol pulses to the Mediterranean area (Bergametti et al. 1989). On the other hand, precipitation scavenging during the rainy season from October to May reduces aerosol concentrations (Dulac et al. 1987). The summer time is characterised by low-inversion layers, and strong sunlight, causing photochemical smog. Moreover, forest fires, which occur during the summer months in the Mediterranean region, increase black carbon and organic aerosol mass as well as fine particle emissions. Thus, the Mediterranean Sea constitutes an area where atmospheric particles originating from continental natural and anthropogenic sources, marine sources and gas-to-particle conversion exist simultaneously. Furthermore, specific meteorological conditions result in high temporal variability of aerosol concentrations. It should be noted that the meteorological conditions in the Mediterranean area are also quite influential for the Central and Southern European weather (Millan et al. 1997; Kallos et al. 1998).

Most of the studies on the chemical composition of Mediterranean particulate aerosol have been conducted in the western and north-western region (e.g. Bergametti et al. 1989; Dulac et al. 1987) or on the eastern coast including Turkey and Israel (Luria et al. 1996; Maenhaut et al. 1999; Koçak et al. 2004). However, relatively few studies have been undertaken in the southern part of the Eastern Mediterranean and Greece (Mihalopoulos et al. 1997; Danalatos and Glavas 1999; Zerefos et al. 2002; Bardouki et al. 2003; Gerasopoulos et al. 2006; Lazaridis et al. 2006).

Notably, there is scarce information concerning continuous aerosol measurement studies to reveal the atmospheric composition and variability of particulate matter and its contribution to air quality and human health in the Mediterranean area. On this basis, continuous aerosol measurements were started during 2003 at the Akrotiri research monitoring station on the western part of the island of Crete (Greece). In the current study the focus is on the ambient levels of PM_{10} and $PM_{2.5}$ and the influence of African desert dust outbreaks on the particulate matter concentrations.

2 Material and Methods

2.1 Site Description

Continuous PM_{10} and $PM_{2.5}$ concentrations were measured at the Akrotiri monitoring station which is situated at an urban background/semi-rural area on the island of Crete, Greece, mid-distance between Athens and the northern African coast at the Akrotiri peninsula. The Akrotiri station (35°31′48″N, 24°03′36″E) is a coastal site (distance from the sea is 2 km for the northern and western directions, 3.3 km for the southern and 11.5 km eastward), approximately 5 km north-eastward away of the city of Chania at an elevation of 137 m from sea level. Therefore, the urban traffic of the city of Chania can affect the concentrations at the station only when the air masses arrive from south-western directions. The region is characterized by a complicated topography (flat surface at the station area, rough rocky surface with small mounds around the station and a small gulch 100 m eastern and 300 m northern from the station; steep rocky bulge at 7 km northern-eastward with five gorges and lowland area where the airport and farmland are situated at 6 km eastward), whereas strong winds, long sunny and humid periods exist. The station was established during 2002 and belongs to the Laboratory of Atmospheric Aerosols of the Department of Environmental Engineering of the Technical University of Crete.

2.2 PM Measurements

Continuous measurements of PM_{10} and $PM_{2.5}$ were performed in time period between 2003 and 2006 (05/02/03–03/09/04 for PM_{10} and 03/10/04–04/02/06 for $PM_{2.5}$) at the Akrotiri research station. However, the July 2003 PM_{10} data and the October–November 2004 $PM_{2.5}$ data are missing due to technical problems in the station. Concentrations of PM_{10} and $PM_{2.5}$ were measured using an automatic beta radiation attenuation monitor (FH 62 I-R Beta gauge, ESM Andersen Instruments GmbH, Erlangen, Germany). In addition to PM measurements, ozone and meteorological data (wind speed and direction, temperature and humidity) were collected on continuous basis. Ambient ozone concentration was measured using Ambient O_3 monitor (APOA 360, Horiba, Japan), while the meteorological conditions were also monitored (wind speed sensor 4034BG; wind direction sensor 4122BG; combined temperature/humidity sensor in shelter 3030.BG; Theodor Friedrichs and Co., Germany).

Simultaneous PM_{10} measurements between the automatic beta radiation attenuation monitor and two manual gravimetric samplers were performed to examine the equivalence of beta monitor results to gravimetric reference method. The gravimetric samplers were represented by a sequential particulate sampler (FH 95 SEQ, THERMO Electron Corporation, Erlangen, Germany) and a standardized gravimetric reference sampler (N.C.S.R. Demokritos, Athens, Greece) equivalent to the reference method according to European standard EN 12341:1998 (CEN 1998). The distance among the sampling inlets of the three above mentioned instruments was 1 meter between each other and the inlets were placed 4 m above the ground level. The measurement was performed during July and August 2006 and it consisted of 32 simultaneous 24-h samplings.

The automatic beta attenuation monitor with a sampling flow rate 1 m^3/h was equipped with PM_{10} head and an automatic heating system. The sampling tube was heated a few degrees above the ambient temperature to prevent the water condensation. Particles were collected on glass fiber filter tape (GF 3516, 40 mm/42 m, Schleicher and Schuell, Germany). Glass fiber filters are known for producing positive artefacts by absorbing acidic acids such as SO_2 and HNO_3.

The sequential particulate sampler consists of a LVS-PM_{10} sampling inlet (at sampling flow rate 2.3 m^3/h), a flow control unit and a storage magazine containing 16 filter cassettes, which was programmed to change the filters automatically every 24 h. The glass fiber filters (GF10, Schleicher and Schuell MicroScience GmbH, Germany) were used for the particle collection.

The flow rate of the gravimetric reference sampler was 1.3 m^3/h and it was controlled by a critical orifice. PTFE membrane filters with 1 μm pore size (Whatman International Ltd., Maidstone, England) were used for the gravimetric reference sampler.

During the continuous measurements of PM_{10} and $PM_{2.5}$, the data from the automatic beta radiation attenuation monitor were collected by a data logger every 15 min and the daily averages were calculated. During the comparative measurement period, the sequential sampler (07/04/06–08/04/06) and the gravi-

metric reference sampler (07/20/06–08/04/06) were operated with a 24-h sampling period having a start time at 15:00 local time. The 24-h averaged values (starting at 15:00) from the beta radiation attenuation monitor were used for comparison with the sequential sampler and the gravimetric reference sampler.

The filters with sampled PM for both gravimetric instruments were stored in closed Petri dishes and equilibrated for 24 h at room temperature and relative humidity of a weighing room before and after sampling. A Sartorius balance (Sartorius CP 225D, Sartorius AG, Goettingen, Germany) with mass resolution of 0.01 mg was used for the weighing of the filters.

It should be noted that the simultaneous measurements were not sufficient for obtaining a correction factor for PM_{10} since further intercomparison with concentrations measured with reference gravimetric equipment must be performed to demonstrate equivalence of beta concentration measurements to reference method (EC 2002). Therefore, the beta attenuation monitor concentrations are presented as measured.

2.3 Back Trajectory Modelling

Trajectories were obtained using the HYSPLIT4 Model (Hybrid Single-Particle Lagrangian Integrated Trajectory), developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (NOAA; Draxler and Rolph 2003; Rolph 2003). The three-dimensional trajectories were computed for the coordinates 35.53N, 24.06E which are the coordinates of the Akrotiri station, at 12 UTC (Coordinated Universal Time) for each day of measurements. The trajectories were 120 h (5 days) long. The wind data were retrieved from NOAA [archived (FNL) meteorological data] and had a time resolution of 6 h and spatial resolution of 191×191 km^2.

2.4 Statistical Analysis

The effect of air masses arriving at the sampling site to PM concentration was examined by performing statistical analysis on the back-trajectory modelling results and the PM concentration data for each day of the measuring period. Four sectors have been used corresponding the north-west (NW), north-east (NE), south-west (SW) and south-east (SE) directions. Each trajectory was divided into 120 segments and each segment has been assigned to one of the aforementioned sectors. Therefore maps for the origin of the air masses were constructed (the maps are presented in section 3.4.2 of the manuscript). The length of each bar in the maps is proportional to the frequency of air masses originating from the specific wind sector as percent (%) of total (vertical axis), whereas the colour corresponds to particulate matter levels of the air originating from that sector (concentration in microgram per cubic meter).

The long range dust transport events were detected in the PM time series. More specifically, we examined the PM time series for potential desert dust events on days with particulate concentration peaks and days having average concentration higher than the monthly averaged value. The prevailing sector for the origin of air masses should correspond to the Southern (Sahara and Sahel deserts) or Western (desert dust event over western Mediterranean) direction for a desert dust event to occur in the area of the station. In addition, for major desert dust events, the concentration peak should be observed also at the urban background/semi-rural Finokalia station, which is located at the eastern part of Crete or at the Heraklion station, located in an urban area close to Finokalia (Gerasopoulos et al. 2006). The back-trajectory modelling results and satellite images from Terra/MODIS were used to confirm the occurrence of a dust event. PM concentrations influenced by desert dust events were subtracted from the dataset and concentrations without dust events were calculated.

In addition, a non-metric Multi Dimensional Scaling (MDS) analysis has been applied to the particulate matter concentration and meteorological data in order to detect and explain the observed similarities between the data for each day of measurements (Clarke and Gorley 2001; Clarke and Warwick 1994). In the MDS analysis the observed similarity matrix between the days of measurements (samples) is analysed and represented in two dimensions. The value of stress shows the extent to which the MDS plot provides a good representation of the similarity matrix of data. The MDS analysis was applied separately for the available PM_{10} and $PM_{2.5}$ concentration data. The similarity matrix was generated, using the Bray–Curtis index of dissimilarity. Then the MDS algorithm was applied and the MDS bubble plots were constructed.

3 Results and Discussion

3.1 Comparative PM_{10} Measurements

In Europe as well as in the US, manual gravimetric methods have been defined as reference methods for particulate matter measurements (CEN 1998; US Government 1991). Therefore, online measurement devices (e.g. beta radiation attenuation monitor) must be examined for equivalence with gravimetric reference methods under various measurement conditions (Salminen and Karlsson 2003; Gehrig et al. 2005).

Comparison between PM_{10} concentrations measured by the beta radiation attenuation monitor, the sequential low volume sampler and the gravimetric reference sampler was performed in the period between 07/04/06–08/04/06 (Fig. 1) in order to examine the equivalence of beta monitor results to gravimetric reference method. The differences between the PM_{10} concentration values given by the online instrument and the two offline gravimetric instruments were within 20% for almost the whole measuring period. The ratio of the beta radiation attenuation monitor concentrations to the sequential sampler concentrations for the whole measuring period was 1.21±0.08, while the ratio between the beta radiation attenuation monitor and the reference sampler was 1.17±0.11. During the period of simultaneous measurements the relative humidity, temperature and wind speed averages were 64.4±8.0% (49.4–78.3%), 25.1±1.9°C (21.8–28.0°C), 2.9±0.9 m/s (1.8–5.9 m/s), respectively. To the range of conditions under which the measurements were conducted the beta attenuation monitor is known of providing concentrations higher than the reference gravimetric method due to water absorption by particles and condensation of evaporation loss from particles on the beta monitor filter (heated inlet to 40°C; Chang et al. 2001; Sillanpää et al. 2002). The beta radiation attenuation monitor filter is not conditioned before the particulate matter mass concentration detection but rather analysed almost immediately when it is sampled. On the other hand, in the case of offline gravimetric instruments, the particulate matter sampling took 24 h and the filters were analyzed up to 16 days (15 days of sampling and 1 day equilibrating the filter in weighing room) after collection for the first filter from the filter magazine of the sequential sampler. Therefore, the higher values measured by the beta radiation attenuation monitor in comparison to the sequential sampler can be caused also by losses on the filters for the gravimetric measurement due to the evaporation of volatile compounds (Saarikoski 2005) and the positive artifacts of the glass fiber filters which are maximized during the summer since SO_2 and HNO_3 concentrations are elevated.

3.2 PM_{10} Measurements

The daily averaged concentrations for PM_{10} during the measuring period from 05/02/03 to 03/09/04 are presented in Fig. 2a. The mean concentration of PM_{10} for the whole measuring period 2003/2004 was 35.0±17.7 μg/m^3 (Table 1). There is a large variability of the daily averaged PM_{10} concentrations mainly during the summer and autumn period with concentrations

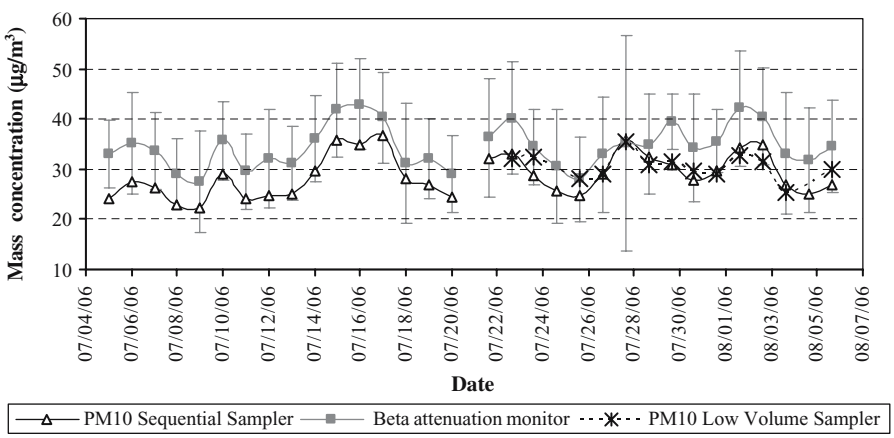

Fig. 1 Comparative measurements of PM_{10} concentrations by the beta attenuation monitor (FH 62 I-R), the low volume sequential sampler (FH 95 SEQ) and the gravimetric reference sampler (N.C.S.R. Demokritos, Athens, Greece). Error bars indicate minimum and maximum counts (±SD)

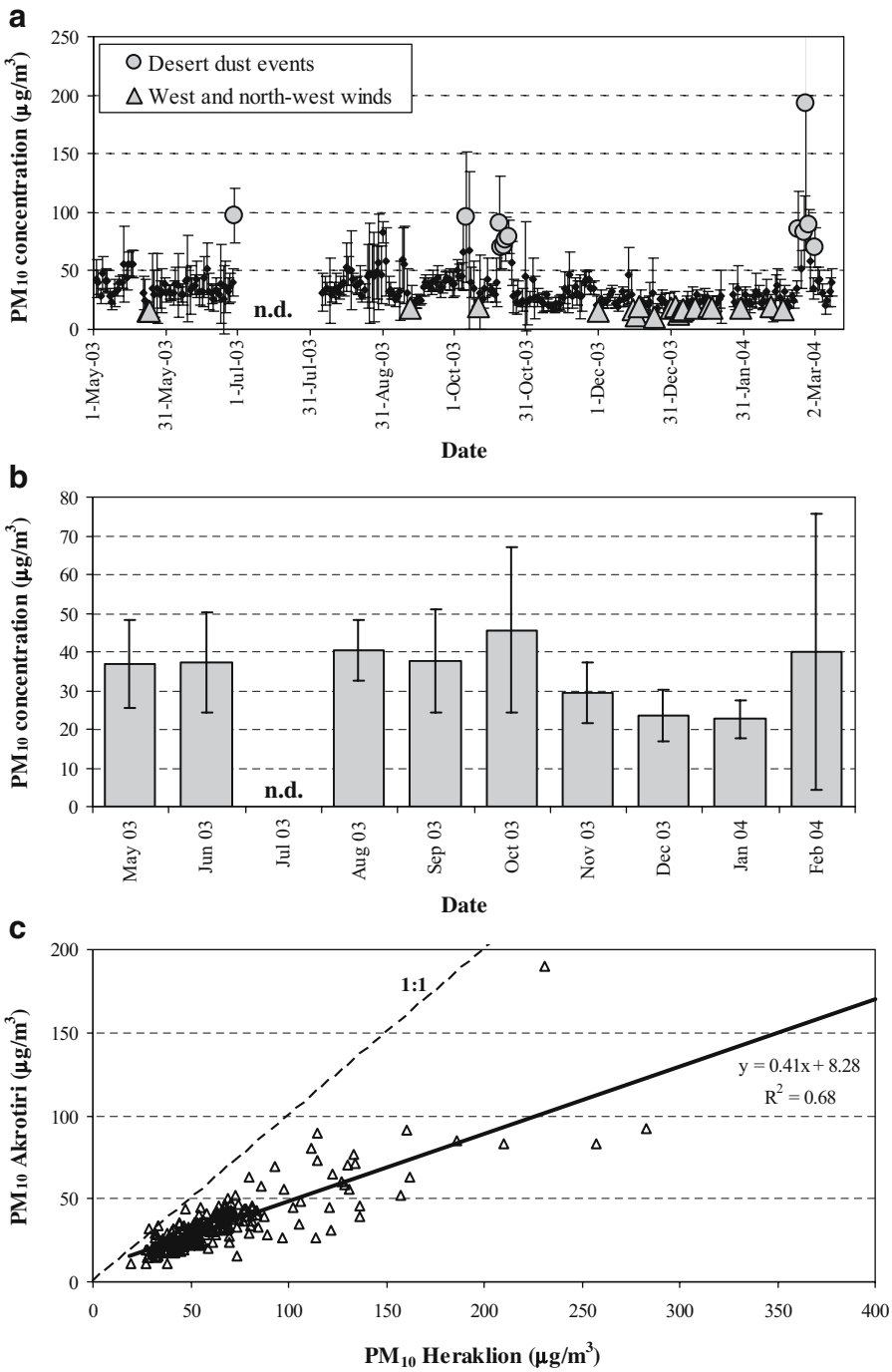

Fig. 2 **a** Daily mean PM_{10} concentrations in the time period between 05/02/03–03/09/04 and **b** monthly mean PM_{10} concentrations during May 2003–February 2004 at the Akrotiri research station by the beta radiation attenuation monitor FH 62 I-R (*n.d.* not determined). Error bars indicate minimum and maximum counts (±SD). **c** Correlation between PM_{10} concentrations at the Akrotiri and Heraklion stations

ranging from 15 to 100 μg/m^3. During the winter period 2003/2004 the daily mean PM_{10} concentrations were in general lower and the variability smaller (26.8±13.7 μg/m^3). However, on the 02/27/04 a major desert dust event lead to an average daily PM_{10} level of 193.2±125.9 μg/m^3. The PM_{10} levels on 02/27/04 between 19:00–21:00 reached more than 400 μg/m^3 with the highest value of 495 μg/m^3 at 20:00, which is a very high concentration even for desert dust episodes (Rodriguez et al. 2002). The days

Table 1 PM_{10} and $PM_{2.5}$ concentration and meteorological parameters statistics at the Akrotiri station

Component	Time period (data capture %)	Averaged value[a]	SD[b]	Min ± SD[c]	Max ± SD[c]
PM_{10} (μg/m^3)	2003/2004 (84)[d]	35.0	17.7	10.0±3.3	193.2±125.9
	2003 (81)	35.7	14.3	10.9±4.1	97.6±23.1
$PM_{2.5}$ (μg/m^3)	2004/2006 (85.4)[e]	25.4	16.5	10.6±3.4	282.4±270.0
	2004 (65.5)	26.1	11.1	11.4±3.9	98.4±95.2
	2005 (95.1)	25.6	17.5	10.6±3.4	282.4±270.0
Temperature (°C)[f]	2003/2004	19.2	5.9	2.1±2.3	32.0±2.7
	2004/2006	18.7	5.8	5.7±0.3	35.9±0.8
RH (%)[f]	2003/2004	70.0	11.8	32.3±2.7	96.4±1.6
	2004/2006	69.4	10.3	29.8±3.5	98.7±1.8
Air velocity (m/s)[f]	2003/2004	3.4	1.7	1.3±0.7	10.8±2.0
	2004/2006	3.7	1.7	1.2±0.7	12.7±3.2

[a] Value averaged over time period

[b] Standard deviation

[c] Based on daily averaged values during the time period

[d] Measuring period for PM_{10} (05/02/03–03/09/04)

[e] Measuring period for $PM_{2.5}$ (03/10/04–04/02/06)

[f] Only for periods with available concurrent PM concentrations.

with major desert dust events are indicated in Fig. 2a (six episodes with duration from 17 h to approximately 5 days and 1 h concentration averages from 60.5–495 μg/m^3; 06/30/03, 10/06/03, 10/20/03–10/24/03, 02/24/04, 02/26/04–02/28/04, 03/02/04). However, the measurements showed 26 temporally short dust episodes (2–6 h) with maximum 1 h averaged concentration of 180 μg/m^3. North Western winds during the measuring period, which in general result to regional diminished particulate matter levels in the observation area, are also indicated in Fig. 2a. The lowest daily PM_{10} value during the measuring period was observed on 02/13/04 (10.0±3.3 μg/m^3) during a storm event. In some cases, high PM_{10} concentrations (e.g. 82.9 μg/m^3 on 09/01/03) were also associated with elevated O_3 levels (corresponding concentration 0.062 ppm) at the station during regional transport from the Central Europe and coincident desert dust event over Crete.

In addition, days with PM_{10} concentration exceeding the 24-h limit value of 50 μg/m^3 are depicted in Fig. 2a. During the period May–October 2003 the limit was exceeded on approximately 20% of the days whereas during the period November 2003–January 2004 no exceedances occurred. In October 2003 (33% of days) and February 2004 (21% of days) the majority of exceedances occurred due to the increased frequency of desert dust events.

In Table 1 averaged values of temperature, relative humidity and air velocity are presented. The PM_{10} concentration values were not correlated with the values of the meteorological parameters. However, PM_{10} concentrations were correlated with temperature (R^2=0.63) when the values influenced by desert dust events were omitted from the dataset.

Figure 2b shows the monthly averaged PM_{10} concentrations during the period May 2003–February 2004. The monthly averaged PM_{10} concentrations without the desert dust episodes are also depicted in Fig. 2b with the semi-continuous line. The July 2003 data are missing due to technical problems in the station (indicated with n.d. in Fig. 2b). Elevated PM_{10} levels were observed in October 2003 and February 2004 with monthly averaged concentrations equal to 45.7±21.4 μg/m^3 and 40.0±35.8 μg/m^3, respectively. The reason for the elevated concentrations during these months was the outbreak of a number of desert dust episodes in the area. Subtracting the desert dust episodes from the data the maximum monthly averaged concentration was observed during the September 2003 and was 36.1±12.2 μg/m^3 (Fig. 2b, semi-continuous line). The lowest monthly mean concentrations were measured during the winter period, in particular from November 2003 to January 2004 (Fig. 2b), when no major desert dust event took place (Fig. 2a).

The annual mean PM_{10} concentration for the year 2003 (based on 8-months measurements) at the Akrotiri station was 35.7±14.3 μg/m^3. Although the levels of PM_{10} at the station are lower than the EU annual limit value for the protection of human health (40 μg/m^3), exceed the EU 2010 limit value of 20 μg/m^3 of PM_{10}. However, PM_{10} concentrations are probably lower than measured according to the results from the comparison with the reference gravimetric method results. The mean concentration of PM_{10} for the whole measuring period 2003/2004 (9-months period; 35.0±12.4 μg/m^3) corresponds to the annual mean concentration of PM_{10} at the urban background/semi-rural Finokalia station, for the year 2004/2005 (28±30 μg/m^3) and at the rural Erdemli station (36.4±27.8 μg/m^3) but are significantly higher than concentrations at rural stations in western Mediterranean (18–30 μg/m^3) and continental Europe (7–25 μg/m^3; Gerasopoulos et al. 2006 and references therein; Koçak et al. 2007 and references therein). Concurrent measurements of PM_{10} at the Finokalia and Akrotiri stations were not taken during the period 2003/2004 and therefore it was not possible to directly compare the evolution of concentrations at the two stations. However, measurements from Heraklion station were available for the period 05/06/03–03/09/04 (63±37 μg/m^3). The PM_{10} concentrations at the Akrotiri station were lower than the concentrations observed at Heraklion station and also were correlated as depicted in Fig. 2c (R^2=0.68).

3.3 $PM_{2.5}$ Measurements

$PM_{2.5}$ measurements were performed from 03/10/04 to 04/02/06 using the automatic beta attenuation monitor equipped with a $PM_{2.5}$ pre-impactor. The daily averages of $PM_{2.5}$ concentrations are shown in Fig. 3a. The average $PM_{2.5}$ concentration during the whole measuring period was 25.4±16.5 μg/m^3. In addition, the annual mean concentrations of $PM_{2.5}$ for the measuring period March 2004–March 2005 and March 2005–March 2006 was 25.5±11.7 μg/m^3 and 25.3±19.2 μg/m^3, respectively. The above concentrations are rather high compared to other rural background stations in the Mediterranean (Koçak et al. 2007 and references therein). On a calendar year basis, the $PM_{2.5}$ annual concentration averages measured at Akrotiri station were 26.1±11.1 μg/m^3 for the year 2004 (based on 10-months measurements) and 25.6±17.5 μg/m^3 for the year 2005 and exceeded the US EPA annual limit value of 15 μg/m^3 and the WHO AQG value of 10 μg/m^3 (WHO 2006). However, as for PM_{10} concentrations, the actual $PM_{2.5}$ concentrations are probably lower than measured.

As for PM_{10} concentrations, there are specific periods during which the daily $PM_{2.5}$ concentration reached high values as a result of desert dust intrusion in the area. Similar observations regarding the effect of desert dust episodes to the $PM_{2.5}$ levels have been reported in the literature (Rodriguez et al. 2004). The days with high $PM_{2.5}$ concentration corresponding to desert dust events and their intensity are depicted in Fig. 3a (03/27/04, 03/28/04, 05/05/04, 05/06/04, 07/12/04, 01/26/05, 01/27/05, 02/15/05, 03/28/05, 04/11/05, 04/17/05, 02/24/06 and 02/28/06). The most intensive desert dust episodes caused a considerable reduction in the visibility (04/17/05, 02/24/06). Especially during the desert dust events of the period January–March 2005 (01/26/05, 01/27/05, 02/15/05 and 03/28/05), the back trajectory analysis showed that the air masses passed over northern Africa close to the ground for many hours (between 36 h and more than 72 h) prior to arriving to the monitoring station. On the 01/26/05 the 24 h averaged $PM_{2.5}$ concentration was 79.5±35.6 μg/m^3 (maximum value 167 μg/m^3), whereas on the 02/15/05 the average 24 h $PM_{2.5}$ concentration reached 54.6±46.1 μg/m^3 (maximum value 215 μg/m^3), on 03/28/05 92.5±93.6 μg/m^3 (maximum value 348 μg/m^3), and on 04/17/05 281.6±270.7 μg/m^3 (maximum 15 min value over 780 μg/m^3).

Figure 3b shows the monthly averaged $PM_{2.5}$ concentration from March 2004 to March 2006. It can be observed from the Fig. 3b that during the winter period (December 2004, January 2005, December 2005 and January 2006) the $PM_{2.5}$ concentrations were lower than during the spring (e.g. April 2005) and the summer period (e.g. July 2005). This trend is in agreement with the PM_{10} measurements and the published PM data for urban background/semi-rural environments in the Mediterranean area (Rodriquez et al. 2002). In Fig. 3b the semi-continuous line corresponds to monthly averaged $PM_{2.5}$ concentrations without the desert dust events. The results show that desert dust events can increase the monthly averaged $PM_{2.5}$ concentrations by a factor of 1.1 to 1.2 during the period from January to May. Especially for April 2005 and February 2006, the value of this factor is 1.5 due to the occurrence of the most intensive desert dust episodes (04/17/05, 02/24/06).

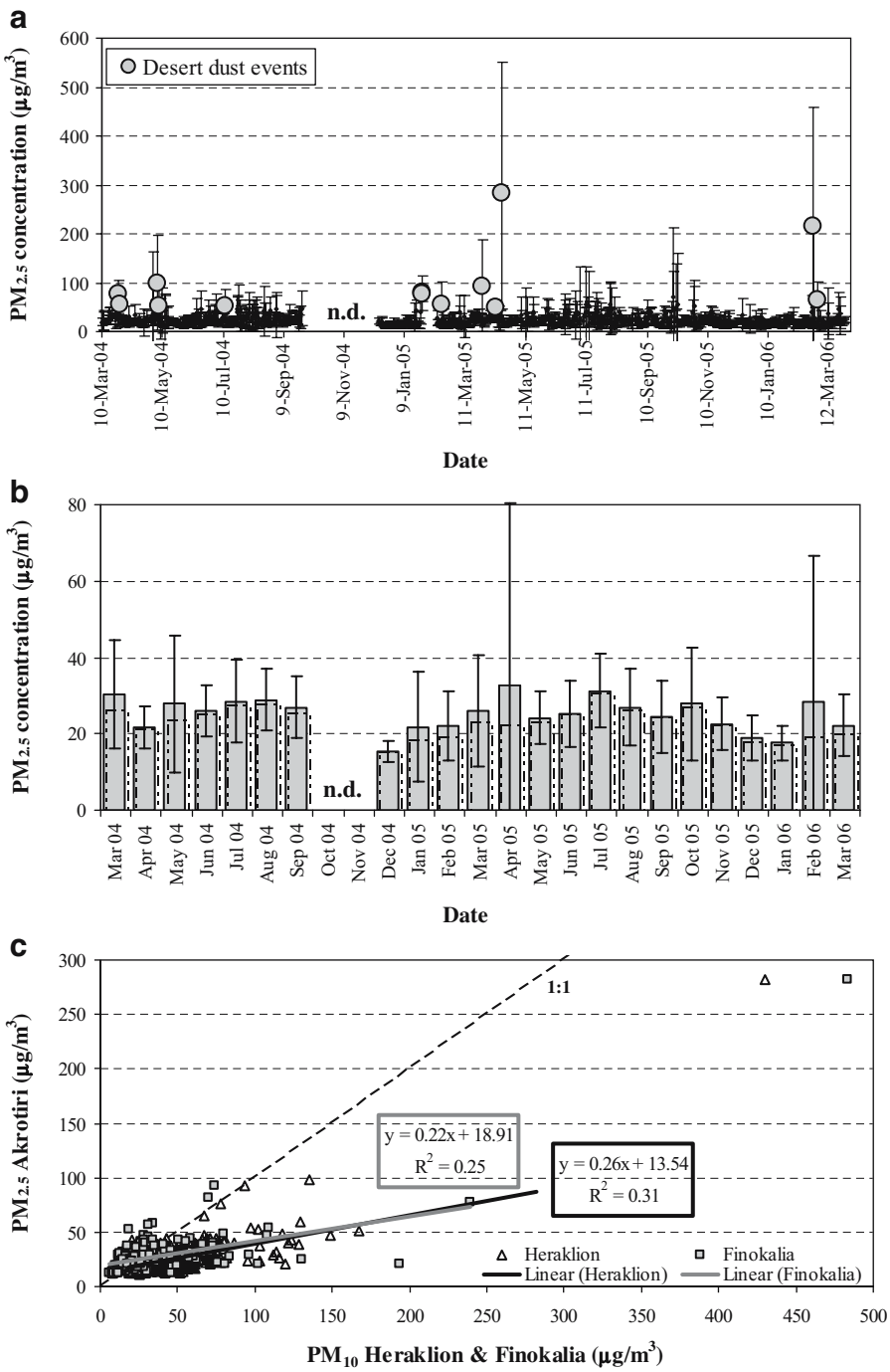

Fig. 3 **a** Daily mean $PM_{2.5}$ concentrations in period between 10/03/04–02/04/06 and **b** mean monthly $PM_{2.5}$ concentrations during March 2004–March 2006 at the Akrotiri research station (Crete, Greece) by the beta radiation attenuation monitor FH 62 I-R (*n.d.* not determined). *Error bars* indicate minimum and maximum counts (±SD). **c** Correlation between PM_{10} concentrations at the Akrotiri and Heraklion stations

Contrary to PM_{10} concentrations, the $PM_{2.5}$ levels at the Akrotiri station were not correlated with the concentrations of PM_{10} at the Finokalia (R^2=0.25) and Heraklion (R^2=0.31) stations during the period 03/10/04–09/30/05 (period with concurrent measurements; Fig. 3c). However, on 04/17/05 elevated PM concentrations were measured at all sites due to the intensive Saharan dust episode. This indicates that sources from the same location, in this case regional transport of Saharan dust from North Africa, contribute significantly to the elevated PM levels in Crete.

3.4 Analysis of the Particulate Matter Concentration Sources

3.4.1 Back Trajectory Modelling

Back trajectory modelling was used to study the origin of air masses arriving to the Akrotiri station and therefore the potential sources of particulate matter concentrations. Representative samples of back trajectory modelling results are depicted in Fig. 4. Figure 4a–b correspond to the measuring period of PM_{10} concentrations, whereas Fig. 4c–e refer to the period of continuous $PM_{2.5}$ measurements at the station. Figure 4a shows an example of back trajectory calculations corresponding to transport of Saharan dust from northern Africa. Specifically, the back trajectory calculations for the 02/27/04, which was the day with the highest PM_{10} concentration in 2004, showed that the air masses passed over northern Africa close to the ground for more than 36 h prior to arriving to the monitoring station. In contrast, in Fig. 5b shows the back trajectory for the 02/13/04, which is the day with the lowest PM_{10} concentration in 2004 (average 24 h PM_{10} concentration 10.0± 3.3 μg/m^3, minimum hourly value 6 μg/m^3). In this case, clean air transport from northern Europe in conjunction with a snow storm in Crete resulted in low PM_{10} concentrations.

Results of the back trajectory analysis for the $PM_{2.5}$ levels at selected dates at the Akrotiri research station are shown in Fig. 4c–e. A Saharan dust episode on 04/17/05 is depicted in Fig. 4c, whereas in Fig. 4d for the 12/05/05 (low 24 h averaged $PM_{2.5}$ concentration of 10.8±2.8 μg/m^3) it is observed that air masses originating from the Atlantic passed mainly above the Mediterranean Sea prior to arriving to the monitoring station. In comparison, Fig. 4e shows that the elevated $PM_{2.5}$ concentration on 07/14/05 (24 h averaged $PM_{2.5}$ concentration: 52.5± 69.9 μg/m^3) were attributed to the regional transport of air masses from the central Europe which passed also from the greater Athens area.

3.4.2 Statistical Analysis of PM Origin

Statistical analysis of the back-trajectory modelling results and the PM concentration data for each day of the measuring period was performed. As regards the PM_{10} measuring period, it was found that the air masses originated mainly from the north with a seasonal variation in frequencies ranging from 78% during autumn to 100% during the summer. In addition, major differences in episodicity were detected, between southern and northern directions. The frequency and length of dust episodes in the area of Akrotiri station were increased when the air masses originated from southern (SW) directions and especially from the Sahara desert. The high aerosol concentrations occurred during specific short time intervals (1–5 days) when southern winds have passed over Africa. When the air masses originated from the NW and NE, the PM_{10} levels were lower. However, elevated concentrations occurred during transport from the north when the air trajectories passed from the Greek mainland and partially from the central Europe.

More specifically, Fig. 5 refers to the origin of air masses and the corresponding average PM_{10} concentrations during the period June 2003–May 2004. During the summer 2003 period [June, July and August (JJA); Fig. 5a] the average PM_{10} concentration levels on days with air masses originating from the NE and NW directions (frequency of occurrence 62.5 and 37.5%, respectively) were equal to 36.8±6.8 and 41.9±14.7 μg/m^3, respectively. Therefore, the regional component from central Europe is the dominant contributor to the particulate matter concentration at the measuring site in summer 2003. During the autumn period in 2003 [September, October, November (SON); Fig. 5b] the PM_{10} concentration levels from the southern directions were higher (SW: 54.9± 22.4 μg/m^3; SE: 49.7±18.4 μg/m^3). However, the frequency of transport from the south is small (approximately 22%) and thus the contribution to the seasonal aerosol concentration is limited (seasonal PM_{10} concentration: 37.7±16.5 μg/m^3; seasonal PM_{10} with air masses originating only from the NE and NW directions: 33.3±11.6 μg/m^3). During winter 2003/2004 [December–January–February (DJF); Fig. 5c] the PM_{10} concentration was low in air masses originating from northerly directions (NW: 25.4± 11.4 μg/m^3; NE: 23.2±4.5 μg/m^3), whereas elevated concentrations occurred due to transport from the south (SE: 35±24.7 μg/m^3; SW: 50.5±48.9 μg/m^3). Finally, Fig. 5d presents the origin of the air masses during spring [March–April–May (MAM)] 2004. It is observed that there was a general increase of the PM_{10} concentration from the northerly directions (NW:

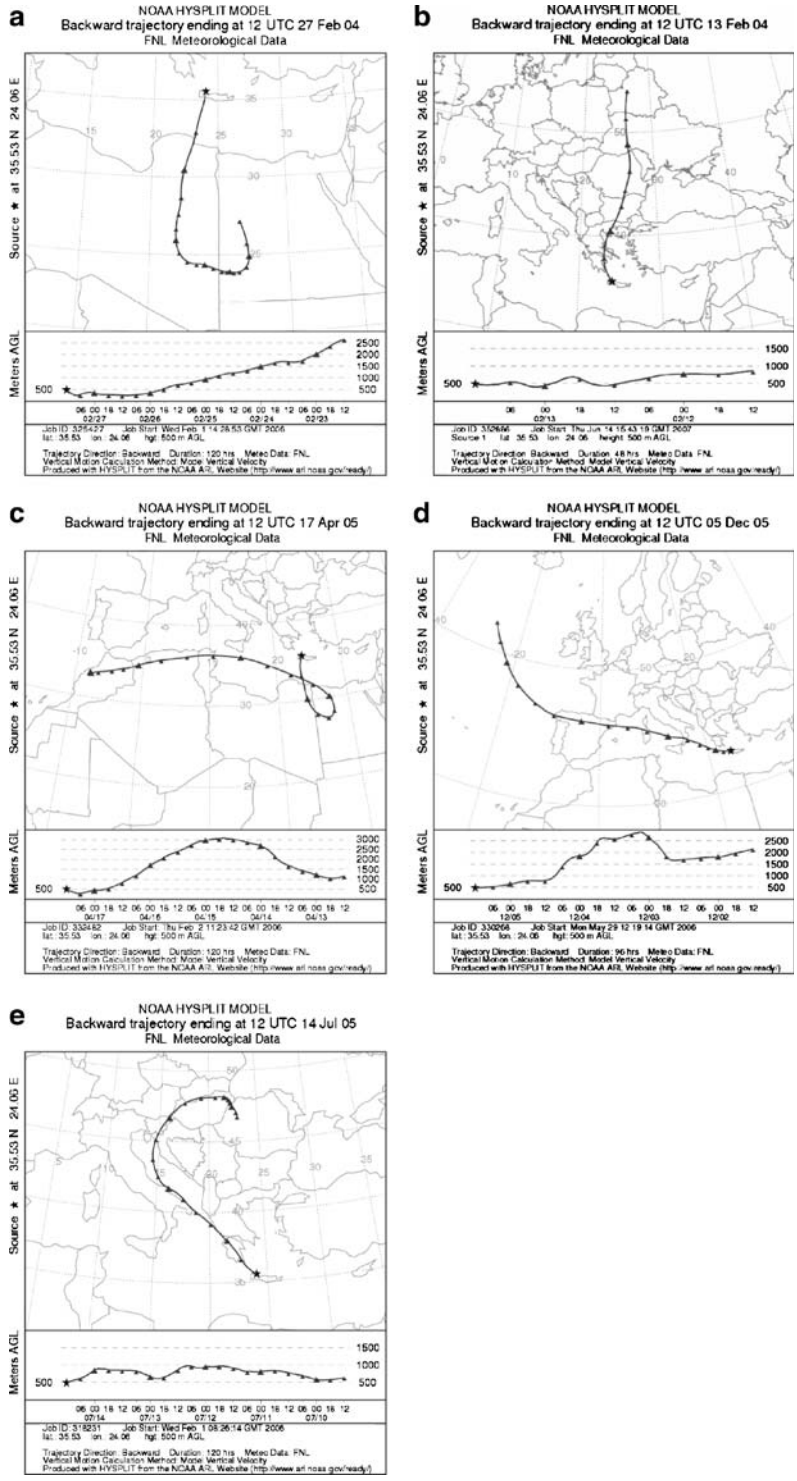

Fig. 4 Back trajectories calculated by the HYSPLIT4 model (Draxler and Rolph 2003; Rolph 2003) for **a** 27th February 2004, **b** 13th February 2004, **c** 17th April 2005, **d** 5th December 2005 and **e** 14th July 2005. In the upper part of the figure a map with the path the air parcel took is presented, and at the lower part a vertical view of its movement at different altitudes (meters above model ground level)

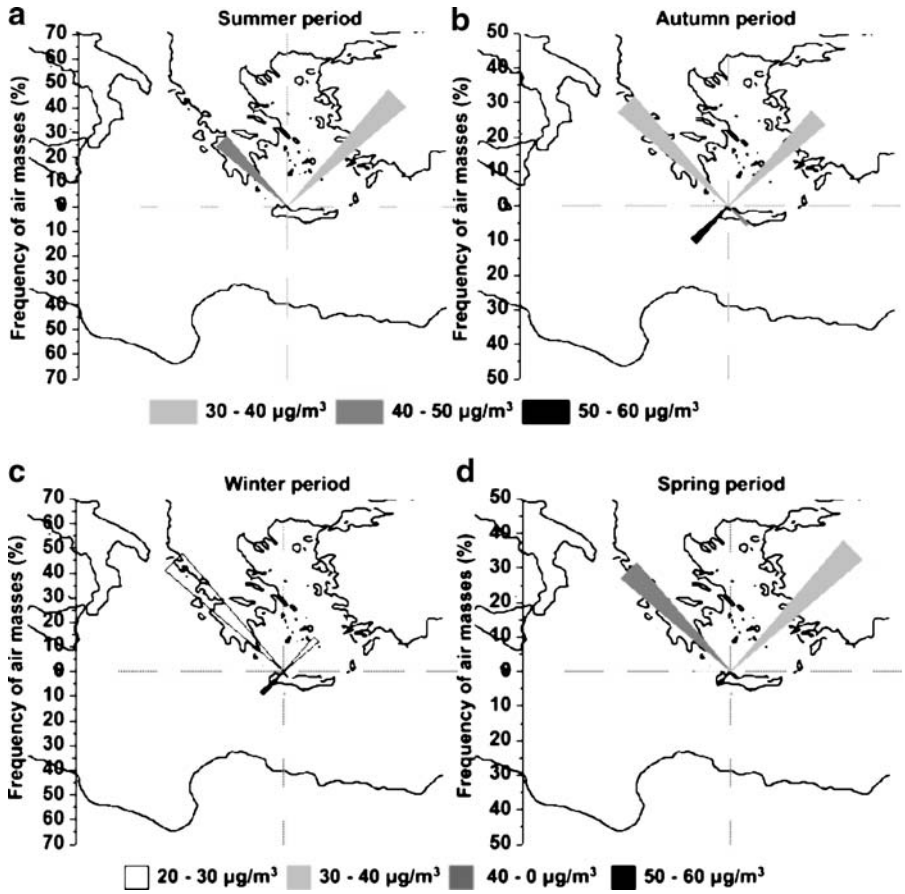

Fig. 5 For the **a** summer 2003 period (JJA), **b** autumn 2003 period (SON), **c** winter 2003/2004 period (DJF), and **d** spring 2003/2004 period (MAM). The vertical axis of the plot shows the frequency of the corresponding air mass origin as percent (%) of total. The colour code shows the concentration in microgram per cubic meter

30.3±9.8 μg/m^3; NE: 39.4±8.3 μg/m^3). Regarding the direction of air masses arriving to the station the most dominant was the NE with frequency of occurrence approximately 50% and the NW with frequency of 41.7%. The air masses originating from the south direction had elevated PM_{10} levels (SE: 55.8 μg/m^3, 2.8%; SW: 55.8±14.4 μg/m^3, 5.5%).

In comparison, Fig. 6a–d present the mean frequency distribution of the $PM_{2.5}$ concentrations at the Akrotiri research station during spring (2004, 2005 and 2006), summer (2004 and 2005), autumn (2004 and 2005) and winter (2004/2005 and 2005/2006) periods, respectively. The comparison of the origin of the air masses and the corresponding average PM_{10} (summer 2003–spring 2003/2004) and $PM_{2.5}$ (spring 2004–winter 2005/2006) concentrations showed significant differences. The average $PM_{2.5}$ concentrations from SW direction were the highest during the spring, summer and winter periods (spring period, SW: 42.1±51.2 μg/m^3; summer period, SW: 34.3±10.1 μg/m^3; winter period, SW: 28.9±35.4 μg/m^3), whereas during the autumn period the highest average $PM_{2.5}$ concentrations originated from NE (autumn period, NE: 27.3±11 μg/m^3). The frequency percentage of the air masses with the highest $PM_{2.5}$ concentrations from SW direction was 12.9% during spring, 3.4% during summer and 22.9% during winter and during the autumn period the frequency percentage of the air masses from NE direction was 54.2%. In contrast, the highest average PM_{10} concentrations were measured from northern directions during summer 2003 (NW: 41.9±14.7 μg/m^3; NE: 36.9±6.8 μg/m^3; frequency 100%).

However, there were some similarities between the origin of the air masses and the corresponding average PM_{10} (summer 2003–spring 2003/2004) and $PM_{2.5}$ (spring 2004–winter 2005/2006) concentrations. The fraction of air masses originating from northerly directions was also for the $PM_{2.5}$ concentrations the most dominant with frequency of occur-

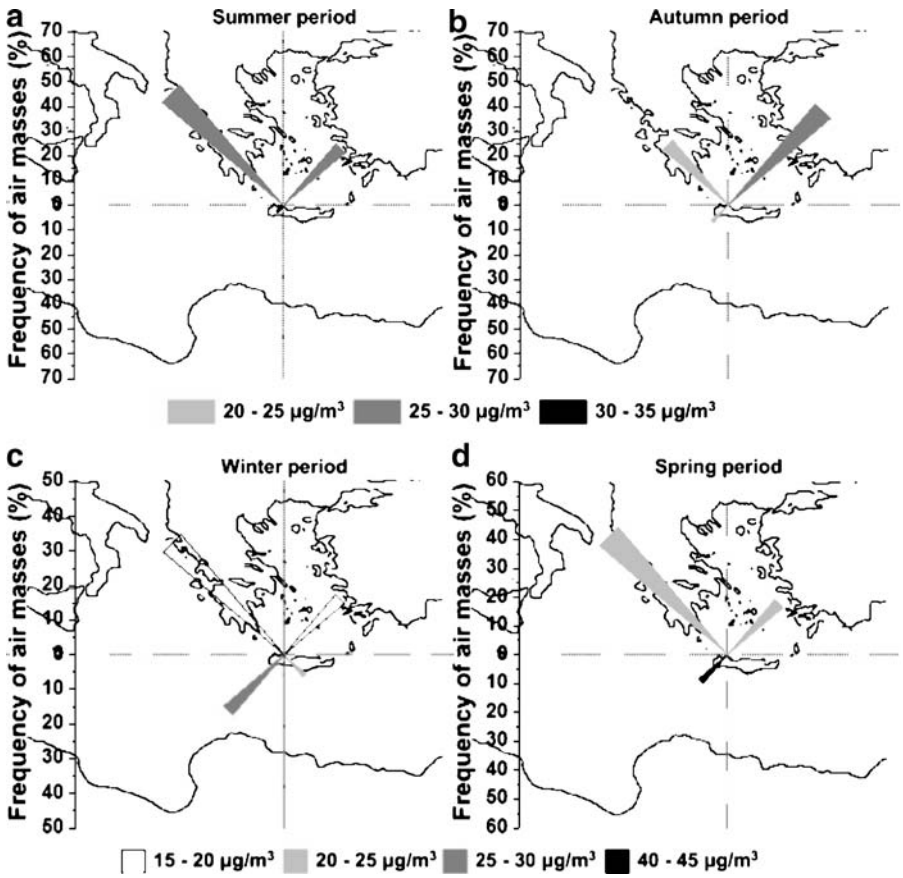

Fig. 6 Origin of the air masses and corresponding average $PM_{2.5}$ concentration (μg/m^3) for the **a** spring 2004, 2005 and 2006 period (MAM), **b** summer 2004 and 2005 period (JJA), **c** autumn 2004 and 2005 period (SON), and **d** winter 2004/2005 and 2005/2006 period (DJF). The vertical axis of the plot shows the frequency of the corresponding air mass origin as percent (%) of total. The colour code shows the concentration in microgram per cubic meter

rence varying from 68.6–96.3%. As for PM_{10} (spring and winter periods of 2003), the $PM_{2.5}$ concentrations originating from southerly directions during the spring and the winter period were in some cases higher (spring period, SE: 27.5±10.2 μg/m^3, SW: 42.1±51.2 μg/m^3, winter period, SE: 23.4±6.6 μg/m^3, SW: 28.9±35.4 μg/m^3) than from northerly directions (spring period, NW: 23.4±8 μg/m^3, NE: 23.5±7.8 μg/m^3; winter period, NW: 17.9±4.9 μg/m^3, NE: 17.8±4.6 μg/m^3). Koçak et al. (2007) have also found that during spring the concentration of PM is elevated in the eastern Mediterranean (Erdemli coastal rural station) due to frequent desert dust events. In addition, on an annular basis the most dominant direction of arrival of air masses at the station was the northern (PM_{10}: 2003, 87.3%; 2004, 80%; $PM_{2.5}$: 2004, 88.1%; 2005; 84.9%; 2006, 74.7%). This is in agreement with wind trajectory analyses in the region performed by Guerzoni et al. (1990) which concluded that more than 60% of air masses which arrive in the eastern Mediterranean come from the N–NW and 13–16% from the Sahara. However, as previously published for the rural Finokalia station in western Crete (Gerasopoulos et al. 2006), the maximum concentrations for both PM_{10} and $PM_{2.5}$ were observed when air masses originated from SW directions (Saharan dust outbreaks; PM_{10}: 2003, 50±22.6 μg/m^3; 2004, 58±49.8 μg/m^3; $PM_{2.5}$: 2004, 31.9±16.6 μg/m^3; 2005, 33±42 μg/m^3; 2006, 34.9±48.3 μg/m^3). The lower concentrations were mainly observed when air masses originated from the NE direction (PM_{10}: 2003, 33.6±9 μg/m^3; 2004, 27.4±11.8 μg/m^3; $PM_{2.5}$: 2004, 24.1±8.3 μg/m^3; 2005, 26.8±10.1 μg/m^3; 2006, 17.9±4.4 μg/m^3).

3.4.3 Non-metric Multi Dimensional Scaling (MDS) Analysis

The MDS analysis results are presented as bubble plots in Fig. 7, separately for the available PM_{10} and $PM_{2.5}$ concentration data. The circles (bubbles) in Fig. 7a and b represent the samples whereas the diameter of each

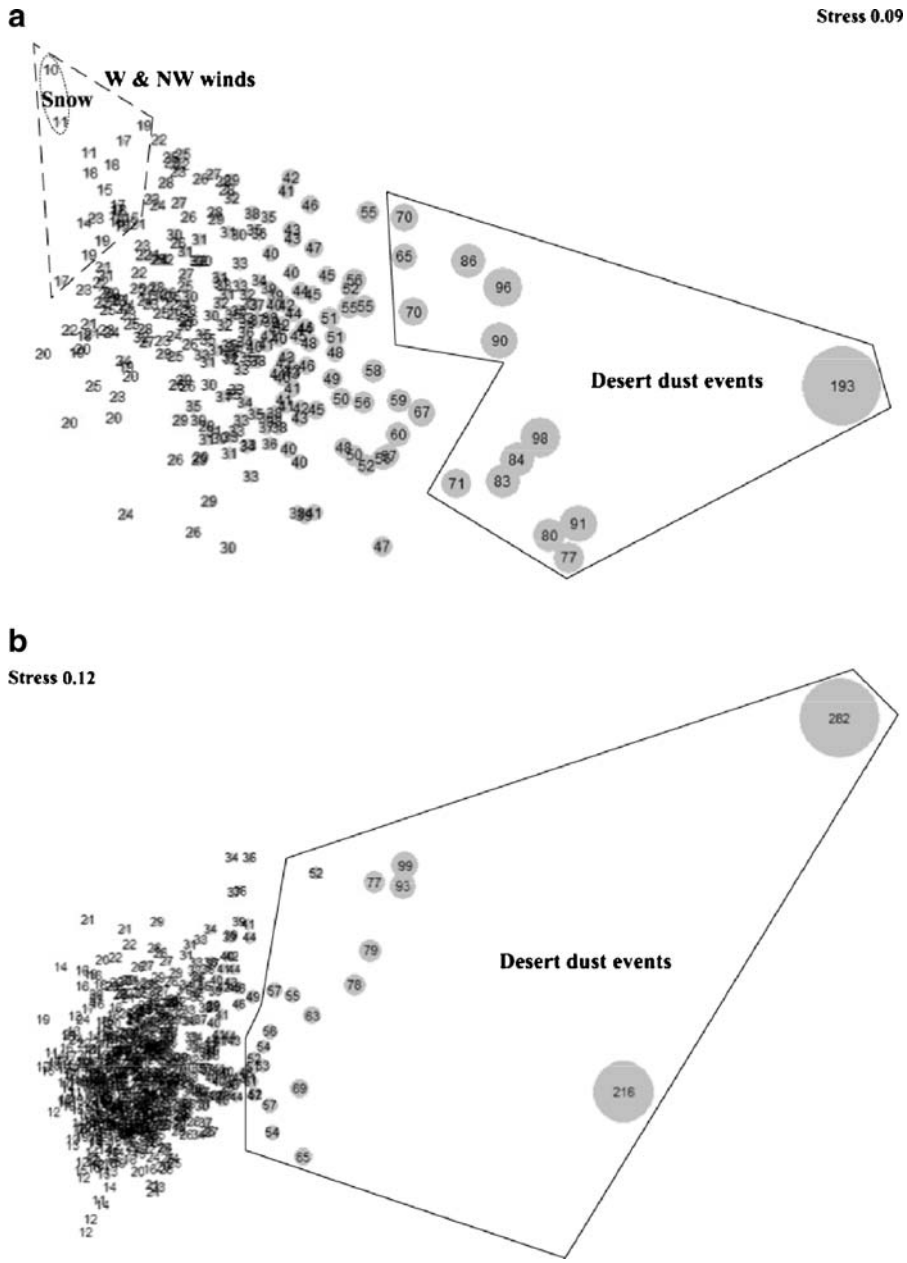

Fig. 7 Non-metric multi-dimensional scaling (MDS) analysis bubble plot for the ambient concentration of **a** 24 h-averaged PM_{10} between April 2003 and March 2004 and **b** 24 h-averaged $PM_{2.5}$ between March 2004 and April 2006, at the Akrotiri research station

circle is proportional to the particulate matter concentration during each day. Bubbles that are grouped together (clusters) represent days with similar characteristics with respect to particulate matter concentration and meteorological parameters. In addition, the representation of the similarity of samples in Fig. 7a and b is satisfactory as depicted by the low stress values (0.09 for PM_{10} and 0.12 for $PM_{2.5}$).

In Fig. 7a where all the 24 h averaged values of PM_{10} are shown, the appearance of three clusters is evident. The right cluster corresponds to high concentrations and is well correlated with desert dust episodes (rare events). The cluster to the left correlates with transfer from western and north-western directions and on two days with snow fall events when the concentrations are very low (rare events). Finally, the middle cluster represents particulate matter concentrations that are most common in the area. We can therefore conclude that the average 24 h PM_{10} values fall in one of three distinctive clusters. The high PM_{10} events

(from 77 to 190 μg/m^3) have similarity with the outbreaks of African desert dust, the low PM events (from 14 to 19 μg/m^3) with the winter storm events (11 μg/m^3), whereas the major part of the data (from 20 to 70 μg/m^3) can be organized in a third cluster where the air masses originate from north-westerly and north-easterly directions. In comparison, the results of the MDS analysis for the $PM_{2.5}$ concentrations are presented in Fig. 7b. Also in this case, most of the $PM_{2.5}$ concentrations fall in a dominant category with values ranging from 12 to 64 μg/m^3, whereas the right cluster represents days with desert dust events (rare events; concentrations ranging from 53 to 282 μg/m^3).

4 Conclusions

Three-year (May 2003–March 2006) particulate matter measurements (PM_{10}, $PM_{2.5}$) using a beta radiation attenuation monitor were performed at the Akrotiri research station on the west part of the island of Crete (Greece) in combination with statistical and back trajectory analysis. The average concentration of PM_{10} for the whole measuring period (May 2003–February 2004) was 35.0±17.7 μg/m^3, whereas the average $PM_{2.5}$ concentration during the whole measuring period (March 2004–February 2006) was 25.4±16.5 μg/m^3. The concentrations of particulate matter undergo a seasonal change characterised by a summer maximum [PM_{10}: 38.7±10.8 μg/m^3 (2003); $PM_{2.5}$: 27.9±8.7 μg/m^3 (2004) and 27.8±9.7 μg/m^3 (2005)] with lower concentrations during winter (PM_{10}: 28.7±22.5 μg/m^3 (2003/2004); $PM_{2.5}$: 21.0±13.0 μg/m^3 (2004/2005) and 21.4±21.9 μg/m^3 (2005/2006)]. In addition, comparative measurements of the PM_{10} concentration between the beta radiation attenuation monitor, a standardized gravimetric reference sampler and a low volume sequential particulate sampler showed that PM_{10} concentrations measured by the beta radiation attenuation monitor were slightly higher than values given by the reference sampler. Especially, the mean ratio of beta radiation attenuation monitor to the standardized gravimetric reference sampler of all results was 1.17±0.11, whereas the mean ratio of beta radiation attenuation monitor to the sequential low volume sampler was 1.21±0.08.

Statistical analysis together with back trajectory studies of the PM data revealed the importance of long range and regional transport contribution to the ambient PM levels at the Akrotiri research station. During the measuring period approximately 15% of air masses arrived at the station from the south (North Africa, Cyprus and South West Asia region on the shore of the Mediterranean Sea) whereas the majority ((NE: 33.6%; NW: 51.2%)) of air masses arrived from the north (Greek mainland and central Europe). The ambient PM_{10} and $PM_{2.5}$ concentrations at the Akrotiri research station show a large variability with elevated concentrations during desert dust episodes [PM_{10}: 93.8±49.1 μg/m^3 (05/02/03–03/09/04); $PM_{2.5}$: 102.9±59.9 μg/m^3 (03/10/04–04/02/06)], whereas low PM_{10} and $PM_{2.5}$ concentration levels are observed during winter when storm episodes and transport from the west Mediterranean Sea and the Atlantic region occur (lower than 20 and 15 μg/m^3 for PM_{10} and $PM_{2.5}$, respectively). Transport from the Greek mainland and the European continent contributes significantly to the particulate matter levels in the area since the prevailing wind direction is northerly throughout the measurement period. The particulate matter concentrations at Akrotiri during transport from northerly direction were elevated (daily averaged concentrations; PM_{10}: ~40 μg/m^3; $PM_{2.5}$: ~33 μg/m^3) but considerably lower than the concentrations during outbreaks of African desert dust.

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