



# Ambient Particulate Matter Concentration Levels and their Origin During Dust Event Episodes in the Eastern Mediterranean

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## Abstract

The eastern Mediterranean region is strongly influenced by long-range transported particulate matter such as desert dust from northern Africa. To investigate the dynamics of Saharan dust events and their origin, satellite images from the MODIS spectroradiometer of NASA's satellites Terra and Aqua, and back trajectories analysis of the HYSPLIT model were combined together with continuous ground-based field data. Field PM<sub>10</sub> and PM<sub>2.5</sub> measurements were performed in the period 2003–2013 at the Akrotiri monitoring station on the island of Crete (Greece). Furthermore, the mineralogical composition of a small number of samples was determined by X-ray powder diffraction, whereas elemental particle composition analysis was performed on the dust samples collected using the ICP-MS technique for a series of elements. The annual average percentage of days with Sahara dust outbreaks in the region of western Crete (Akrotiri station) was 9.3% for the period 2003–2013. Excluding the PM<sub>10</sub> exceedances caused by Sahara dust events, the average PM<sub>10</sub> monthly concentrations are decreased by 20.5% during the 8-month period November–June, with higher PM<sub>10</sub> concentration reduction (29.9%) in the period February–April. Mineralogical analysis showed that illite was the most abundant mineral identified in all samples, followed by quartz and calcite. Gypsum was detected only in the dry sample. Moreover, the elemental particle composition analysis showed that collected dust originated from Africa deserts.

**Keywords** Sahara dust events · Dust transport · Eastern Mediterranean · Greece · PM mass concentration

## 1 Introduction

Transport and deposition of atmospheric air-borne dust in the European continent is largely determined by the surface dust emissions from Africa and the movement of air masses. In fact, the Sahara Desert, the largest source of dust particles in the world, is estimated to emit about 220 million tons of dust annually, which is transported to the atmosphere by induced winds with considerable consequences for the global climate, the ecosystems and the human health (Moreno et al. 2006; Rodriguez et al. 2015). In general, mineral dust, originated from desert regions of the world, contributes considerably to the air-borne particle load. The Sahara and Sahel arid regions stretch over an area of around

3000 km<sup>2</sup>, but there is not the same emission load of particulate matter from all the regions. Saharan dust particles are composed mainly of oxides (e.g., SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, FeO, Fe<sub>2</sub>O<sub>3</sub>, CaO) and carbonates (CaCO<sub>3</sub>, MgCO<sub>3</sub>)—principle constituents of the Earth's crust (Vanderstraeten et al. 2008).

Emitted particles from wind erosion can be transported over long distances such as the Mediterranean Basin, the Atlantic Ocean and Scandinavia (Rodríguez et al. 2001; Febo et al. 2010). The island of Crete is one of the regions that are more influenced by Aeolian dust originated from the African deserts since it is located just 300 km north from the African coast. Indeed, air-borne African dust can reach Crete in a course of only few hours (Nihlén et al. 1995).

Saharan dust is resuspended and transported via the major synoptic systems (Griffin et al. 2001; Kellogg et al. 2004; Jenkins et al. 2017). The resuspended particles undergo mobilization into the atmosphere by strong surface winds and convection that creates vertical turbulent mixing, and then carried over long distances (Schütz 1980; Wang and Liu 2014; Gkikas et al. 2016; Marinou et al. 2017).

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During Sahara dust events, ambient PM concentrations increase significantly and the inhalation of high PM concentrations can cause serious problems to human health (Prospero 1999; Moreno et al. 2006 and references therein; Goudie 2009; Pandolfi et al. 2014; Mandija et al. 2017). Some studies concluded that bioaerosols, like pollen, viruses, fungi and bacteria, can be attached to the dust transported (Griffin et al. 2001; Kellogg and Griffin 2003). Moreover, African dust can affect the climate, both directly and indirectly (Rosenfeld et al. 2001), as well as the creation of clouds (Gerasopoulos et al. 2009). Additional impacts include changes on atmospheric chemistry and changes on the chemical composition of oceans and forests (Guieu et al. 2002), by enriching them with nutrients.

The overall contribution of transported African dust to the PM<sub>10</sub> levels in the Mediterranean area can be significant and African dust may represent up to 45% of PM<sub>10</sub> concentration levels in the area (Rodriguez et al. 2002; Escudero et al. 2007; Pey et al. 2013; Aleksandropoulou and Lazaridis 2013).

The current study aims to examine the origin of the air-borne particles during dust events in the region of Crete, to identify and analyse the dust events that took place during the period 2003–2013 and to determine the mineralogical and elemental particle composition of a small number of air-borne Sahara samples which were collected during the period 2004–2009.

## 2 Sampling Site and Methods

The sampling site (Akrotiri monitoring station) is an urban background/marine monitoring station on the island of Crete (Greece), about 240 km from the Greek mainland (Athens) and 320 km from the northern African coast (Kopanakis et al. 2012). A detailed description of Akrotiri station is given by Lazaridis et al. (2008) and Kopanakis et al. (2012).

Continuous PM measurements (PM<sub>10</sub>/or PM<sub>2.5</sub>, using the corresponding PM inlets) were performed with an automatic beta radiation attenuation monitor (FH 62 I-R, THERMO Electron Corporation, Erlangen, Germany). PM concentrations were recorded at 15-min intervals. Meteorological data were also collected on continuous basis during the total sampling period (Lazaridis et al. 2008).

In the current study, a daily data analysis was performed to determine dust transport episodes for the period 2003–2010 and 2013. The aforementioned years were selected because there are available data for the majority of the days. In addition, during the current work, five air-borne dust samples were collected in canisters in spring of 2004, 2005, 2006 and 2009. All samples were collected during days with intense Saharan dust episodes (low visibility). The samples were divided into two categories, according to the method of collection

and the type of dust deposition. The deposited dust samples from rainfall residues were collected during or after a rain event, whereas the deposited dry dust sample was collected via an impactor on a canister. The rain samples were filtered to remove rain water, dried at 105 °C and stored in plastic bags. The mineralogical composition of these different air-borne Saharan dust samples was also determined.

An objective of the current study is to identify the characteristics of Sahara dust episodes (origin, frequency, PM concentration levels) observed at the Akrotiri monitoring station. In order to investigate the origin of the particles, satellite images from NASA's Moderate Resolution Imaging Spectroradiometer (MODIS) instrument, and back trajectories analysis of HYSPLIT4 model (Hybrid Single-Particles Lagrangian Integrated Trajectories, Draxler and Rolph 2003) were used. For the days with elevated particulate matter concentrations the HYSPLIT4 (NOAA) model was used in order to determine the origin of air masses (backward trajectory analysis).

The mineralogical composition of the samples was determined by X-ray powder diffraction (XRD). The X-ray powder diffraction method (XRD) was used to characterize the crystallographic structure, the crystallite size, and the preferred orientation in polycrystalline or powdered solid samples. The data were obtained at 35 kV and 35 mA, with a graphite monochromator, using CuK $\alpha$  radiation. A 0.02° scanning step and 1 s counting time, per step, were used for the range 3°–70°. Since all samples were powdered, dust samples minimum grinding was applied. The data were evaluated qualitatively with the Software Diffrac Plus (SOCABIM SAS France). The quantitative analysis was carried out with the Rietveld Method (Bish and Howard 1988) using the software Autoquan.

Finally, the elemental particle composition analysis of the dust samples collected at Akrotiri research station was performed with the ICP-MS (Inductively Coupled Plasma Mass Spectrometry) technique for a series of elements such as Si, Al, Ca, Fe, Mg, K, Cu, Zn whereas, ratios of the relative abundance of the different elements with respect to Si were computed to determine their origin. The elements were determined by ICP-MS after aqua regia digestion of the samples in sealed PTFE bottles at 120 °C. All reagents used were of high purity and the standard reference material GXR-2 was analyzed along with the samples in order to check the reliability of the method.

## 3 Results and Discussion

### 3.1 Variations of Particulate Matter Concentrations During Sahara Dust Events in the Region of Crete

In the region of Eastern Mediterranean, the probability of elevated PM levels is higher compared to Western

Mediterranean due to the higher occurrence and intensity of African dust intrusions (Querol et al. 2009; Nastos 2012; Kopanakis et al. 2015). At Akrotiri station, several Sahara dust events were observed during the period 2003–2009 (Lazaridis et al. 2008; Kopanakis et al. 2012). The dust events mainly occurred during late winter and spring, as for instance the major outbreaks on the 27th of February 2004 (max value of  $PM_{10}$  at  $495 \mu\text{g}/\text{m}^3$ ), the 27th of March 2004 ( $PM_{2.5}$   $121 \mu\text{g}/\text{m}^3$ ), the 9th of February 2005 ( $PM_{2.5}$   $745 \mu\text{g}/\text{m}^3$ ), the 18th of May 2005 ( $PM_{2.5}$   $143 \mu\text{g}/\text{m}^3$ ), the 23rd of March 2008 ( $PM_{10}$   $119.6 \mu\text{g}/\text{m}^3$ ) and several other episodes. During these dust events, huge quantities of Saharan and Sahelian dust were transported to the Mediterranean basin and to the area of interest of the current study. The duration of the Sahara dust events observed at Akrotiri station varied a lot and ranged from few hours to 2 days. Time-series of daily PM mass concentrations during the period 2003–2013 are depicted in Fig. 1. Several daily values of  $PM_{10}$  or  $PM_{2.5}$  (according to the PM inlet used at each measurements period) are higher than  $100 \mu\text{g}/\text{m}^3$ .

In this study, the daily limits of the European Community (1999/30/EC and 2008/50/EC directives) for  $PM_{10}$  ( $50 \mu\text{g}/\text{m}^3$ ), the mean annual target value of EU and the daily limit of WHO guidelines for  $PM_{2.5}$  ( $25 \mu\text{g}/\text{m}^3$ ) were used. The Directive 2008/50/EC on ambient air quality and cleaner air for Europe provides Member States with the possibility to subtract the contribution of natural sources under certain conditions before comparing the ambient air pollutant concentrations to the limit values. Transport of natural particles from dry regions is considered a natural source (Jones et al. 1995; Prospero 1999).

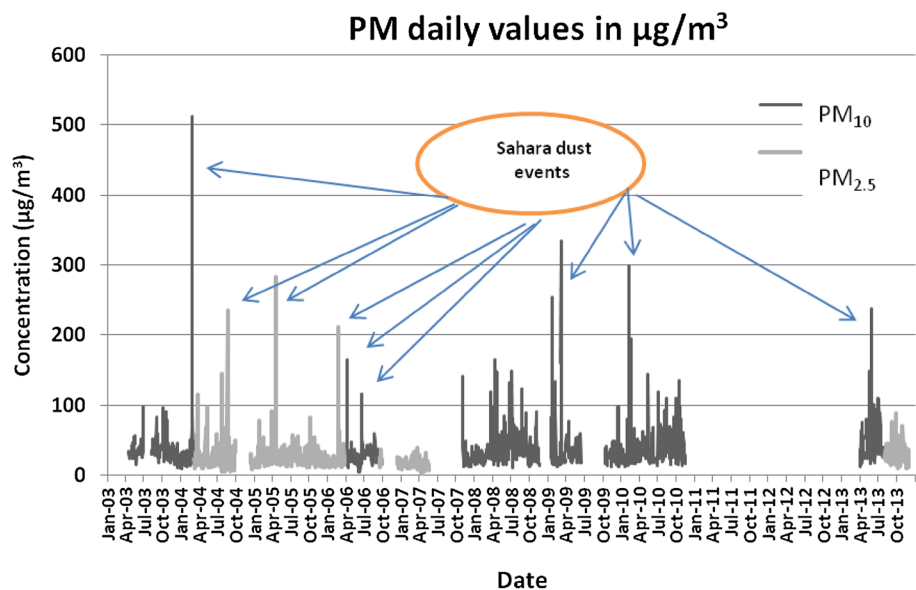
In Table 1, the number of days with an occurrence of Sahara event during the 8-month cold (October–May) or 4-month warm (June–September) season for the years

2003–2010 and 2013 (for 2011 and 2012 there are no available data for the whole year) are presented. On average, the data coverage for those 8 years was 72.8%. A proportion of about 10% of the Sahara dust events occurred in the period June–September (summer season), while the main part, about 90% during the 8-month colder period, October–May (winter season). As it is presented in Table 1 only 21 from the 222 Sahara event cases are recorded during the warm period. During the summer northeast winds, known as *etesians* (or *meltemia*), blow in the island prevailing the transportation of Sahara dust (Kopanakis et al. 2015).

In Table 2, the number of exceedances of the daily  $PM_{10}$  and  $PM_{2.5}$  limits for each month of the years 2005, 2008 and 2010 (the 3 years with the broader data coverage were chosen, on average 89.2% coverage) at Akrotiri station is shown.  $PM_{2.5}$  measurements were conducted in 2005, while in 2008 and 2010  $PM_{10}$  data were recorded. As it can be observed from both Tables 1 and 2, the majority of exceedances of the  $PM_{10}$  and  $PM_{2.5}$  limits occurred in the period January–May. The majority of Sahara dust episodes occurred from February to May, whereas during September and October fewer events were observed. This observation is in agreement with other studies (Pey et al. 2013). In this study the shortest Sahara dust event lasted 2 h (30/06/2008) whereas the longest about 78 h (26–29/02/2004), durations that are in agreement with other Saharan dust observations in central Mediterranean (Meloni et al. 2008).

In 2005 ( $PM_{2.5}$  measurements, data coverage 95%), 134 exceedances of the daily limit took place, most of them during the summer months. However, only 14 of the 134 exceedances were due to African dust transport. The rest of the  $PM_{2.5}$  exceedances can be attributed to local particle sources (Kopanakis et al. 2012). In 2008 ( $PM_{10}$  measurements, data coverage 89%) 67 exceedances of the daily limit

**Fig. 1** Time-series of measurements (daily values) over the years 2003–2013 for  $PM_{10}$  and  $PM_{2.5}$  at the Akrotiri station



**Table 1** Number of days with Sahara event during the 8-month cold (October–May) and the 4-month warm (June–September) season for the years 2003–2010 and 2013 at Akrotiri station, Greece

Year	2003	2004	2005	2006	2007	2008	2009	2010	2013	Sum
Number of sampling days (% data coverage)	211 (57.8)	282 (77.0)	347 (95.1)	279 (76.4)	202 (55.3)	326 (89.1)	207 (56.7)	304 (83.3)	234 (64.1)	2392 (72.8)
Days with Sahara event (cold season)	20	15	29	9	15	40	26	31	16	201
Days with Sahara event (warm season)	1	0	0	7	0	11	0	0	2	21
SUM	21	15	29	16	15	51	26	31	18	222

of  $50 \mu\text{g}/\text{m}^3$  were recorded (Table 2), 39 of which can be attributed to Sahara dust transport. In 2010 ( $\text{PM}_{10}$  measurements, data coverage 83% of days), 74 exceedances of the daily limit were recorded. Half of those cases (37) were due to dust transport from the region of Sahara. During the period October–May (Table 2), almost all the daily  $\text{PM}_{10}$  exceedances were due to African air-borne dust, unlike the period June–September, when the main reasons of  $\text{PM}_{10}$  exceedances were associated with emissions from local sources (Kopanakis et al. 2012).

Table 3 shows the contribution of Saharan dust events to monthly exceedances of  $\text{PM}_{10}$  concentrations in 2008 and 2010. During the period October–May, a high percentage of the daily  $\text{PM}_{10}$  exceedances (23 out of the 30 exceedances during 2008, 32 out of the 34 exceedances during 2010) was due to African dust events. In contrast, in the period June–September, exceedances occurred mainly due to local anthropogenic activities. In total, during 2008 and 2010 41.8 and 50.0% of the exceedances were attributed to Saharan dust episodes. Furthermore, there were several cases of Sahara dust transport where  $\text{PM}_{10}$  concentrations remained below the daily limit value of  $50 \mu\text{g}/\text{m}^3$ .

In Fig. 2a, b the monthly  $\text{PM}_{10}$  (Fig. 2a) and  $\text{PM}_{2.5}$  (Fig. 2b) concentrations, calculated both by taking and not taking into consideration the contribution of Sahara dust events are presented. After excluding the contribution of Sahara dust transport, the largest decrease in the average monthly concentrations is observed during the 8-month period November–June. Regarding  $\text{PM}_{10}$  concentrations the highest reduction observed in February, March and April with a reduction of 17.6, 8.0 and  $9.9 \mu\text{g}/\text{m}^3$ , respectively (42.7, 22.6 and 24.4%, respectively). If the elevated  $\text{PM}_{2.5}$  concentrations due to Sahara dust events are not taken into consideration, the average monthly concentration decreases significantly in February, March, April, September and October (16.3, 10.5, 16.5, 15.5 and 18.2% respectively). The largest variability in the average monthly concentrations is observed during the cold 8-month period, which can be attributed to local particle sources such as vehicular traffic and wood combustion for heating.

In addition, higher reductions in the monthly average concentrations were observed in the periods, when  $\text{PM}_{10}$  measurements were conducted. Therefore, the influence of Saharan dust is more evident for coarse particles in the region of Crete, where mostly coarse particles are transported from the deserts of North Africa under certain meteorological conditions.

Mean African dust contributions to  $\text{PM}_{10}$  (in  $\mu\text{g}/\text{m}^3$  and as percentage) were calculated for both 2005 and 2008. The African dust contribution is obtained by calculating the difference between the experimental  $\text{PM}_{10}/\text{PM}_{2.5}$  concentration value and the calculated 40th percentile value (Escudero et al. 2007; Pey et al. 2013). This is one of the official

**Table 2** Seasonal exceedances of the daily PM<sub>10</sub> and PM<sub>2.5</sub> limit for the years 2005, 2008 and 2010 at Akrotiri station, Greece

Year Month	2005 (PM <sub>2.5</sub> )		2008 (PM <sub>10</sub> )		2010 (PM <sub>10</sub> )	
	IND. <sup>a</sup>	S.E. <sup>b</sup>	IND.	S.E.	IND.	S.E.
Jan	2	5	0	0	0	2
Feb	2	4	0	0	0	8
Mar	8	6	0	3	0	3
Apr	11	5	1	10	0	4
May	10	1	4	3	1	3
Jun	15		10	1	2	1
Jul	21		10	2	11	1
Aug	14		7	1	14	1
Sep	11		5	1	8	2
Oct	11	2	0	2	1	6
Nov	9	1	2	5	0	3
Dec	6	5	0	0	0	3
Sum	120	29	39	28	37	37
June–September	61	0	32	5	35	5
October–May	59	29	7	23	2	32

<sup>a</sup>Number of exceedance events—dependent of dust transport<sup>b</sup>Number of exceedance events—dependent of dust transport (Sahara effect)**Table 3** Statistical analysis per month for the years 2008 and 2010, indicating the involvement of air-borne dust from Africa on exceedances of PM<sub>10</sub> recorded in Akrotiri station

(a)			(b)		
2008 (21/11/2007–20/11/2008)			2010 (15/11/2009–14/11/2010)		
Month	Days with C <sub>PM<sub>10</sub></sub> > 50 µg/m <sup>3</sup>	Saharan dust events days	Month	Days with C <sub>PM<sub>10</sub></sub> > 50 µg/m <sup>3</sup>	Saharan dust events days
January	0	0	January	2	2
February	0	0	February	8	8
March	3	3	March	3	3
April	11	10	April	4	4
May	7	3	May	4	3
June	11	1	June	3	1
July	12	2	July	12	1
August	8	1	August	15	1
September	6	1	September	10	2
October	2	2	October	7	6
November	7	5	November	3	3
December	0	0	December	3	3
Total number of days (362)	67	28	Total number of days (350)	74	37

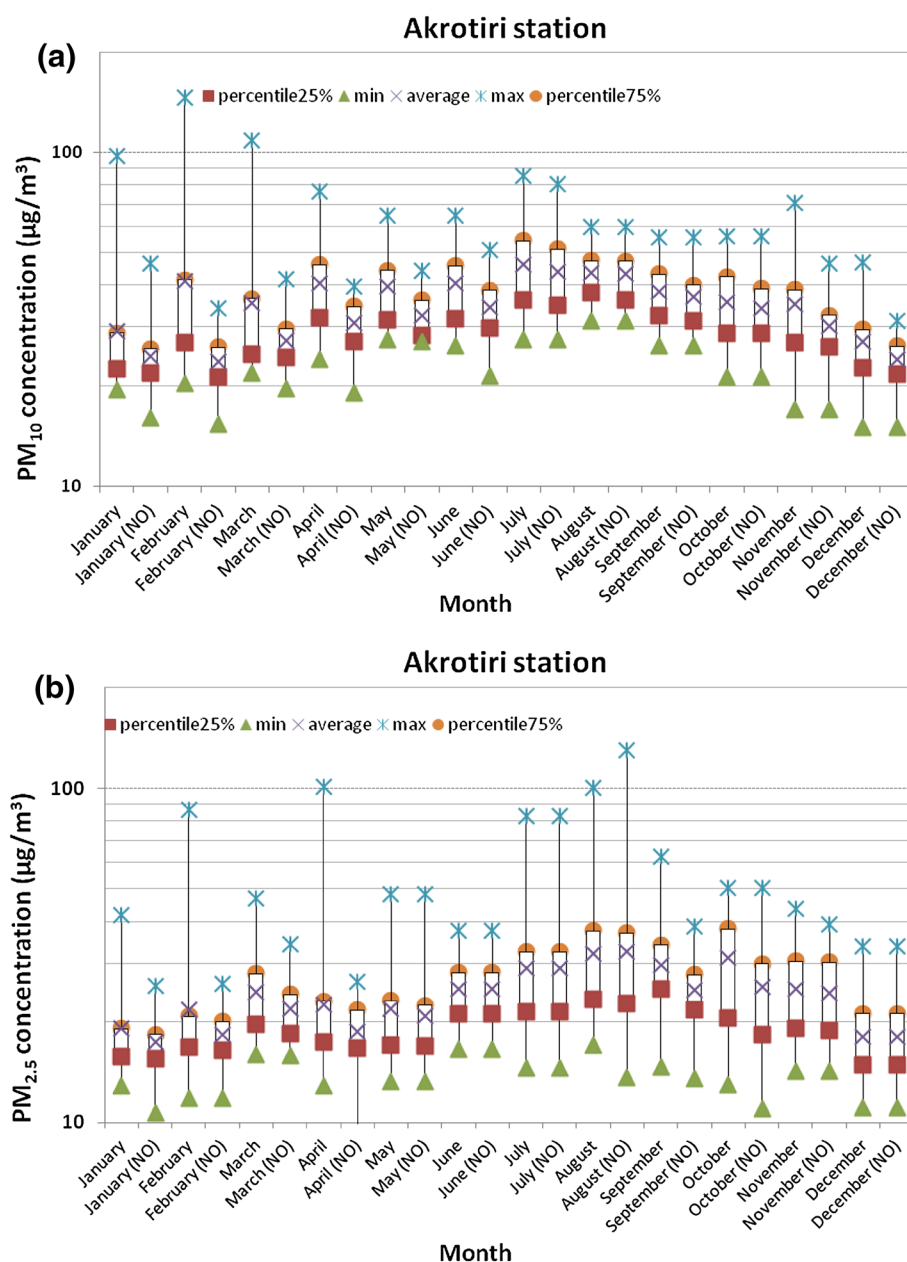
methods adopted by the European Commission for evaluating the occurrence of African dust outbreaks and quantifying its contributions as discussed in the European Guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC (2008) on ambient air quality and cleaner air for Europe.

The African dust contribution for 2005 was about 5.2 µg/m<sup>3</sup> (19.5%) in PM<sub>2.5</sub>, whereas the contribution of African air-borne dust to PM<sub>10</sub> (2008) was 10.4 µg/m<sup>3</sup> (23.3%). The, respectively, African dust contributions to PM<sub>10</sub> were 7.3 µg/

m<sup>3</sup> (35.0%) and 8.4 µg/m<sup>3</sup> (35.0%) at Finokalia and Cyprus stations, respectively (Pey et al. 2013). The above results show that the area of eastern Crete was affected more by intensive dust storms originated from North Africa, than from Middle East or Negev deserts.

In total, during the period 2003–2013 (with data coverage 72.8%) 9.3% of the measurement days were influenced from Sahara dust episodes in the region of western Crete, where 91% of them were recorded during the 8-month winter period October–May. During the studied period, there

**Fig. 2** Mean monthly concentrations and statistics results for **a**  $PM_{10}$  and **b**  $PM_{2.5}$  calculated with and without the contribution of the Sahara dust events



was no evidence of changing trends on African dust contribution to ambient  $PM_{10}$  levels, only sporadic annual peaks as a result of some intensive episodes.

### 3.2 Origin and Transport of Saharan Dust Events

Back trajectories analysis with the HYSPLIT4 model provided the origin sources of the dust reaching Akrotiri station. Most frequent source of origin was Libya with Algeria and Tunisia following. The air masses before reaching Akrotiri station frequently traveled above more than one country with Libya–Tunisia–Algeria and Libya–Egypt being the

most frequent paths. In a small number of cases, the origins of the dust carrying air masses were Morocco or Mauritania.

Moreover, five dust samples were collected in 2004, 2005, 2006 and 2009 during Sahara dust events (Table 4). The days that the five samples were collected south winds prevailed in the area. Transported air masses stayed at least for 1 day above the African deserts before moving north to the island of Crete. The air-borne dust resulted in elevated  $PM_{10}$  concentrations. As an example, high  $PM_{10}$  concentrations were observed on the 17th of April 2005 (with a mean daily value:  $282.4 \mu\text{g}/\text{m}^3$ ) and on the 5th of March 2009 (mean daily value:  $160.1 \mu\text{g}/\text{m}^3$ ) (the next day—6th March 2009, a higher mean value was recorded:  $334.1 \mu\text{g}/\text{m}^3$  and a max

**Table 4** Mineralogical composition (in wt%) of the eolian dust samples

Sample	Date	Illite	Quartz	Calcite	Kaolinite	Dolomite	Palygorskite	Albite	Chlorite	Gypsum
S1 (rain)	5/5/04	30.0	19.3	21.2	8.2	5.8	6.3	9.3	–	–
S2 (rain)	17/4/05	29.8	19.0	28.7	4.8	4.7	4.9	7.4	–	–
S3 (rain)	21/4/06	38.6	15.0	17.0	5.1	3.2	3.5	12.0	4.6	–
S4 (rain)	5/3/09	28.7	25.8	12.7	6.9	4.3	3.5	14.0	4.5	–
S5 (dry)	5/3/09	30.0	24.5	13.0	8.0	3.5	3.0	10.0	3.0	5.0
Samples from rain (average)	–	31.8	19.8	19.9	6.3	4.5	4.6	10.7	2.3	–
Difference (dry-rain samples)	–	– 1.8	4.7	–6.9	1.7	– 1	– 1.6	– 0.7	0.7	5.0

value of  $PM_{10}$ :  $667 \mu\text{g}/\text{m}^3$ ) when two of the samples were taken.

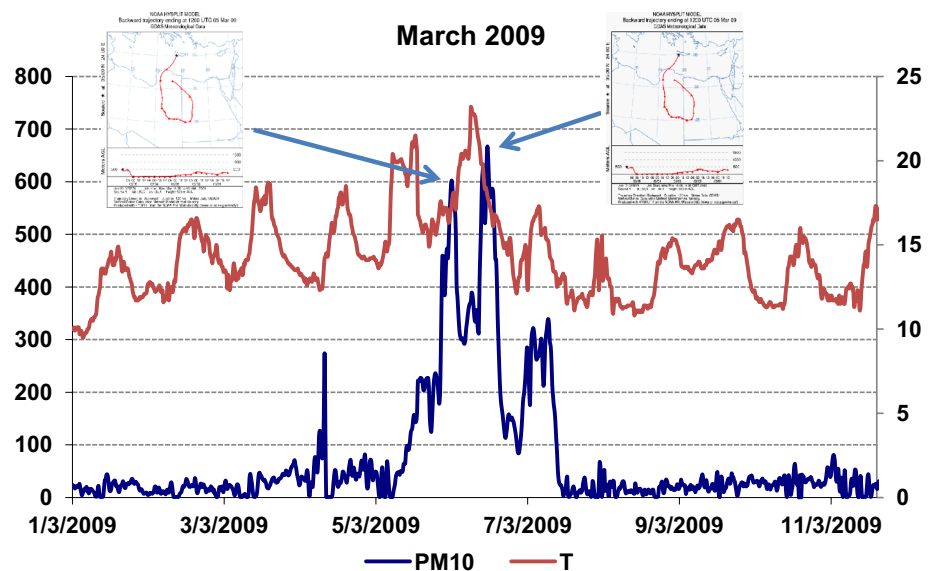
Figure 3 shows the time-series correlation of temperature and  $PM_{10}$  concentrations between the 1st and the 13th of March 2009 when a major Sahara dust event occurred (on 5th–8th March 2009). It is observed that between 1st and 6th March, the temperature was increasing gradually, whereas a simultaneous increase of air-borne particle concentration was observed, mainly after the 5th March. This temperature increase can be ascribed to the transport of hot southern air masses originated from the Saharan region of North Africa. To confirm the southern origin of air masses and to investigate the path of air parcels before reaching the island of Crete, back trajectories of the air masses were derived using the HYSPLIT model. The derived back trajectories showed the North African long-range transport pattern and that the air masses were originated from the regions of Egypt and east Libya (Libyan desert) (Fig. 3).

Figure 4 shows the back trajectory of the air masses arrived to Crete on the 17th April 2005 when a wet dust sample was collected. The origin of the air masses proves that the elevated concentrations can be attributed to Sahara

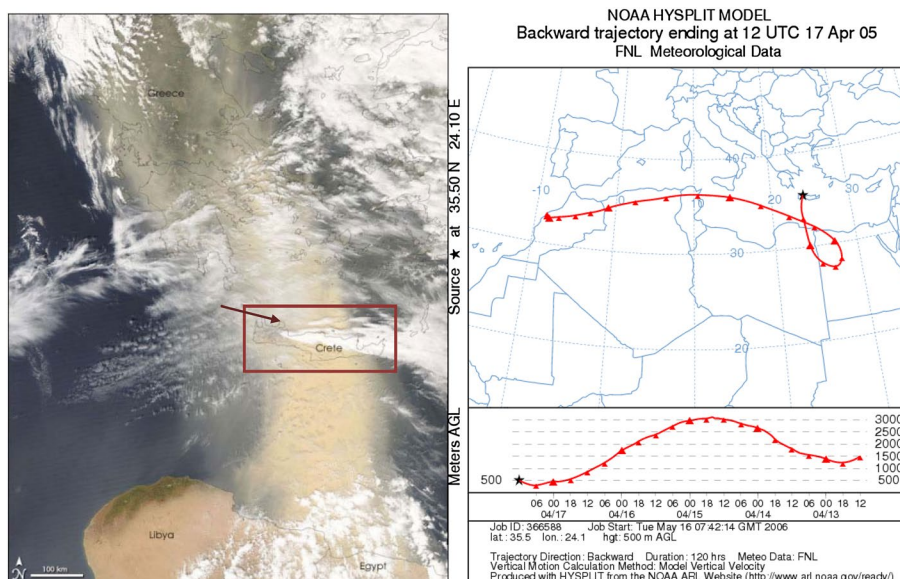
dust transport. As it is observed, the air masses 5 days before reaching Crete initially passed above the Atlas Mountain and afterwards they passed above Sahara and Sahel areas further enriching the air masses with dust particles. Figures 3 and 4b show the difference in the altitude at which the air parcel moved during these days. While before the Sahara dust event, that took place on the 17th April 2005, the air-borne particles moved at high altitude (2 days before arriving at Crete were at 3000 m aloft), at the Saharan dust event on the 5th of May 2009 the air masses moved at low altitude close to the sea level (see Fig. 3). When air masses are moving at low altitude, particles can be more easily deposited to the sea surface. Furthermore, when examining the trajectory, on 6th March 2009 (Fig. 3), a totally different route of the air masses can be seen. The increased mean concentration of air-borne dust measured on the 6th March in relation to the dust concentrations on 5th March can be attributed to the higher altitude at which the air mass moved that day.

The transport of Sahara dust in the Mediterranean region has been studied also by satellite images. Figure 5 shows MODIS satellite images for 3 days when rain samples were collected at the Akrotiri station (see also Table 4). In the first

**Fig. 3** Time-series of  $PM_{10}$  concentrations and atmospheric temperature during the period 01/03/2009–11/03/2009. The y axis refers to  $PM_{10}$  particle mass ( $\mu\text{g}/\text{m}^3$ ) and the mean daily temperature values ( $T \times 20$ , in  $^{\circ}\text{C}$ ). Inside the figure are shown the 5-day-back trajectories of air masses on 5th and 6th March, 2009, at the 500 m' arrival height



**Fig. 4** **a** MODIS satellite image of dust transport episode on 17th April, 2005 (NASA). That day the wet sample S2 was collected, **b** Southern back-trajectory drive dust to Crete on 17th April 2005. The red rectangular shows the area of interest (Crete) and the arrow the Akrotiri monitoring station (color figure online)



satellite image (Fig. 5a), the layer of African dust is clearly seen and its forefront part is covering the region of the island of Crete. While in the other two photos (Fig. 5b, c), a dense cloud is covering the area of western Crete, where the station of Akrotiri is located. Furthermore, the analysis on the days with intense Sahara dust incursions showed the presence of low-pressure systems in the west coasts of Europe and the north Tyrrhenian Sea. Strong surface and mid troposphere winds that transported significant amount of dust over Greece contributed to the Sahara dust transport.

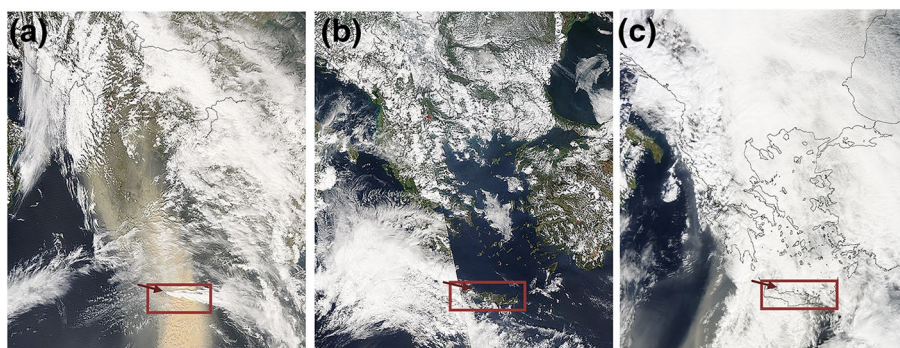
The driving force of the whole phenomenon seems to be an anticyclone over the northwestern Africa, which transports desert particles over the central and eastern Mediterranean with the air masses following an anticyclonic pathway. During the transportation of particles, much of the initial particle load cannot reach distances such as the island of Crete due to deposition on the sea surface (Querol et al. 2009; Nastos 2012).

The back-trajectory analysis which was performed during the period 2003–2013 showed that 90.2% of air masses, originated from north Africa, reaching the Akrotiri station have moved at altitudes equal or lower than 2000 m,

whereas on the rest 9.8% cases the air masses came from altitudes higher than 2000 m. The masses coming from 2000-m altitude and below need on average 2.5 days to approach Crete, whereas when they come from higher altitudes require 3 days. The particle concentrations observed in the area of interest on both circumstances are similar (the average concentrations for the period 2003–2013 were 71.5 and 71.2  $\mu\text{g}/\text{m}^3$ , respectively). The cases where air masses are originated from altitudes above 4000 m are few (about 1.7%), whereas the cases that air masses reaching Crete are originated from heights lower than 500 m are about 26.4%. Therefore, the majority of air masses arriving to the island of Crete from south direction are originated from altitudes lower than 2000 m in the north Africa, where they are enriched with dust particles.

The above conclusion is in accordance with other studies regarding the region of eastern Mediterranean (Querol et al. 2009; Nastos 2012). These studies showed that over the eastern Mediterranean, most of the African dust affecting ground-based measurements was transported at surface levels. However, the transport of Sahara dust towards the

**Fig. 5** MODIS (AQUA, NASA) satellite images above the region of Greece taken on **a** 17th April, 2005 **b** 21st April, 2006 and **c** 5th March, 2009. The red rectangular shows the area of interest (Crete) and the arrow the Akrotiri monitoring station (color figure online)





western Mediterranean mostly occurs at relatively high atmospheric levels (Escudero et al. 2005).

The total annual number of Sahara outbreaks in the region of west Crete was lower compared to the number of Sahara dust events in Cyprus and in Finokalia (eastern Crete) (Pey et al. 2013). The average annual percentage of elevated particle concentration due to Sahara outbreaks in the region of Akrotiri station was 9.3% for the period 2003–2013, whereas at Cyprus and Finokalia were 34 and 31%, respectively (Pey et al. 2013). These regions are situated eastern than Akrotiri station and they are more affected by dust outbreaks from Middle East and Negev deserts.

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 (Pey et al. 2013). The African storms in the eastern Mediterranean from spring to early summer, which lead to the transport of heavily loaded air masses, are associated with the west–east motion of Sharav cyclones (Meloni et al. 2008).

### 3.3 Mineralogical Analysis

The particulate material of the Sahara–Sahel region is mainly constituted by quartz, calcite, dolomite and clay minerals. Direct analysis of Ca, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, K, Mg, and the indirect determination of Si ( $3 \times \text{Al}_2\text{O}_3 = \text{SiO}_2$ ) and CO<sub>3</sub><sup>2-</sup> ( $1.5 \times \text{Ca} + 2.5 \times \text{Mg} = \text{CO}_3^{2-}$ ) allows the calculation of the mineral load contributed by the Saharan region (EC 2011; and references therein).

Table 4 shows the samples specification, the dates of sampling, and their mineralogical composition whereas Table 5 presents the concise chemical form of the measured minerals during the field measurements at the Akrotiri station. As it is presented in Table 4, illite was the most abundant mineral identified in all samples. The mean percentage was 31.8% in the rain-collected samples and 30.0% in the air-borne dust sample. Those results are in agreement with other studies determining the mineralogical composition of dust samples originated from the Northern Africa (Blanco et al. 2003; Menéndez et al. 2013). Ganor and Foner (1995) showed that the percentage of illite is higher when dust is transported from the regions of Chad, compared to the cases the air masses are transported from the Egyptian and Libyan deserts.

Illite, quartz and calcite were the main components of mineral dust as observed in our study, in accordance with other XRD measurements on desert-derived dust (Blanco et al. 2003). Furthermore, kaolinite showed an average value equal to 8.0% in the rain-collected samples and 6.3% in the air-borne dust sample.

**Table 5** The chemical composition of the measured mineral dust particles

Mineral	Chemical form
Illite	$\text{K}(\text{Al}, \text{Mg}, \text{Fe})_2(\text{Si}, \text{Al})_4\text{O}_{10}[(\text{OH})_2(\text{H}_2\text{O})]$
Quartz	$\text{SiO}_2$
Calcite	$\text{CaCO}_3$
Kaolinite	$\text{Al}_2(\text{Si}_2\text{O}_5)(\text{OH})_4$
Dolomite	$(\text{CaMg})(\text{CO}_3)_2$
Palygorskite	$(\text{Mg}_5, \text{Al})(\text{Si}_4\text{O}_{10})_2(\text{OH})_2 \cdot 8(\text{H}_2\text{O})$
Albite	$\text{Na}(\text{AlSi}_3\text{O}_8)$
Chlorite	$(\text{Mg}, \text{Fe})_3(\text{Si}, \text{Al})_4\text{O}_{10}(\text{OH})_2(\text{Mg}, \text{Fe})_3(\text{OH})_6$
Gypsum	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$

Moreover, there are differences between concentrations in air and rain samples (Table 4), which suggest that calcite presents higher relative abundance in samples collected during rain than those of dry air. In contrast, the rest of the minerals (except gypsum) show similar percentages in both liquid and dry samples. The only exception is the gypsum which has the tendency to dissolve in rain-water samples and remains unaffected in dry sample where shows an abundance of 5% (Table 4). Rain may also be an important factor in altering soil crust compositions (Rashki et al. 2013).

Smectite is a mineral-index for air parcels originating from the central Algerian region. The lack of smectite in our samples denotes that air masses in Crete do not originate from Algerian dust sources for the specific number of samples measured in our study. Furthermore, Ganor and Foner (1995) found that smectite is a major (55%) element of dust originated from Libyan and Egypt regions. Smectite, a scarce mineral, can be met also in hyper-arid or semi-arid areas from Northern China and Greenland to North Africa. However, the number of samples in our study was limited and no certain conclusions can be drawn.

Minerals show different apportionments in deserts around the world. For instance, Saharan- and Sahelian-derived eolian dust frequently contains small quantities of chlorite (in our study close to 2.3%) less than kaolinite, whereas in Asia chlorite can be met at higher concentrations than kaolinite, a significant mineral of African dust (Rashki et al. 2013). Moreover, albite and palygorskite, which constitute 10.6 and 4.3% of the sample, respectively, do not exist in the list of the first seven abundant minerals in deserts of North China. Potential sources of palygorskite are areas of Western Sahara and southern Morocco (Molinari 1996).

In Table 6, the differences between the ratios illite/kaolinite (I/K) and kaolinite/chlorite (K/Ch) in the aerosol samples collected at Akrotiri station are presented. These two ratios are important because they indicate the origin of particles. The ratio K/Ch can be used as a good proxy for tracing the dust origin on a regional scale. Low K/Ch ratio (below than

**Table 6** Diagnostic ratios between minerals at the collected eolian dust samples at Akrotiri station

Sample	Date	Illite/kaolinite	Kaolinite/chlorite
S1 (rain)	5/5/04	3.7	–
S2 (rain)	17/4/05	6.2	–
S3 (rain)	21/4/06	7.6	1.1
S4 (rain)	5/3/09	4.2	1.5
S5 (air)	5/3/09	3.8	2.7
Average (SD)		5.1 (1.7)	1.8 (0.8)

1) show that the dust is originated from an Eastern Asian source. On the other hand, Saharan and North American dusts are characterized by abundance of kaolinite compared to chlorite, leading to higher K/Ch ratios (ranging from 1 to 10) (Caquineau et al. 2002). In fact, as it is shown on Table 5, all collected dust samples, were originated from the North Africa region and contained higher quantities of kaolinite than chlorite (ratios between 1.1 and 2.7), with a mean K/Ch ratio value equal to  $1.8 \pm 0.8$ . The value of the K/Ch ratio in combination with the air mass back-trajectory routes could adequately be used as a method of identification of the air dust origin.

Additionally, the relative abundance of illite and kaolinite has been used to trace the regional origin of Saharan dust (Caquineau et al. 2002). The higher the I/K ratio was, lower ratio of K/Ch was observed. In fact, deserts of north China show low K/Ch ratios and higher I/K ratios, whereas in the region of north Africa and Mediterranean basin higher values of K/Ch ratio (average  $\approx 8$ ) and lower for I/K ratio were recorded (5.1 in the current work).

### 3.4 Elemental Particle Composition Analyses

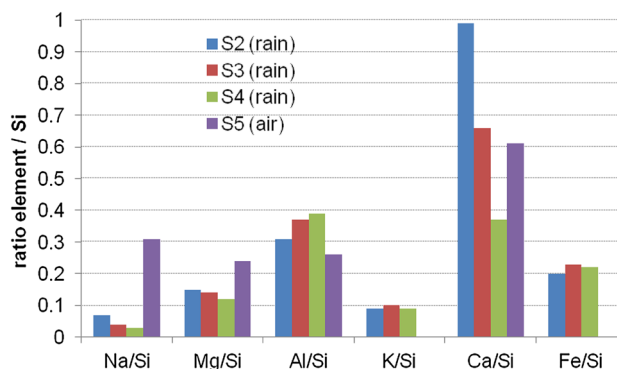
Elemental particle composition analysis was also performed on the dust samples. This analysis was performed for four samples, three wet (S2: 17/04/2005; S3: 21/04/2006; S4: 05/03/2009) and one dry sample (S5: 05/03/2009). The ICP-MS analysis showed that all dust samples contained a significant amount of elements such as Si, Al, Ca, Fe, Mg, K and Na, whereas traces of several other elements as Cu, Zn, B, Mn, Ni, Cr, V and Ba were detected. The ratios of the relative abundance of the different elements with respect to Si have been calculated since Si has a relatively constant concentration in the earth's crust and in addition, its concentration is affected to a limited extent by the human activities. Figure 6 presents the ratios of the main elements with respect to silica.

The Al/Si ratio is a good factor to determine the differences between desert- and anthropogenic-dominated samples, with higher values observed in samples originated from deserts and lower ones in anthropogenic samples (Blanco

et al. 2003). Al/Si ratios higher than 0.3, generally show that dust samples are derived from deserts (Guerzoni et al. 1997). In this work, the values of Al/Si ratio, especially for wet samples from rainfall residues, are close to 0.3 (Fig. 6) (mean value of the four wet samples equal to 0.33). The above values suggest that the collected dust was originated from African deserts. In the dry dust sample there is absence of some main (K, Fe) and trace elements (Mn, Sr, Hg, U). In fact, in Fig. 5 the air sample collected on the 5th of March 2009, is characterized by a lack of potassium (K) and iron (Fe). This finding can be ascribed to the lack of smectite.

Furthermore, several minerals or/and other chemical species dissolve in water of the rain and so, they were not detected in the wet sample. There is a notable difference between concentrations of main elements in the rain-derived samples and in the dry air-borne dust. In fact, these differences (the percentage of the dry to wet samples), were calculated (average value of three rain-dust samples), and they were 5.6, 7.2, 7.8, 14.2 and 54.3% for Al, Ca, Si, Mg and Na, respectively, with average difference 17.8%. In conclusion, concentrations of the main elements of minerals are lower in the case of the dry-collected sample. This implies that the air-borne dry particles contain higher percentage of organic and inorganic compounds and lower amount of earth-born minerals, and that a considerable amount of ingredients, such as organic compounds and soft minerals (such as gypsum, smectite) are soluble in the water of rain. As an example, gypsum, a mineral composed of calcium sulfate dihydrate, is moderately water soluble (2.0–2.5 g/L at 25 °C). The solubility of gypsum in water was also confirmed from the XRD analysis, where gypsum was not detected in the wet samples.

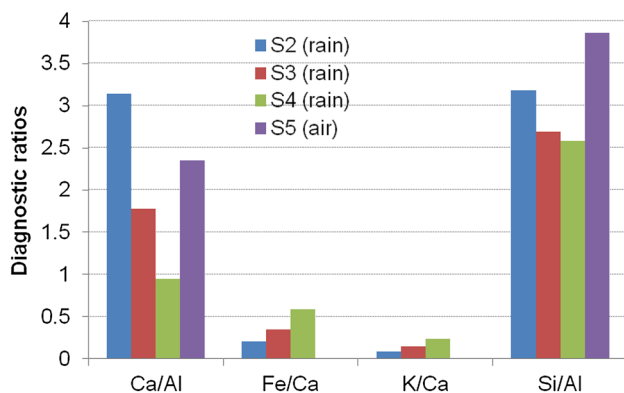
The Ca/Si ratio showed very high values ranging from 0.37 to 0.99, values significantly higher than the value stated in the study of Blanco et al. (2003), where the ratio ranged from 0.10 to 0.15. High Ca/Si, Ca/Al and Si/Al ratios can



**Fig. 6** Element/Si ratios of concentration (for the elements: Na, Mg, Al, K, Ca and Fe) of the collected Saharan-derived dust (wet and dry) samples at Akrotiri station, Crete

be ascribed to dust samples with a larger content of quartz, calcite and dolomite. Moreover, the dust originated from northwestern Sahara (mainly from Moroccan Atlas) is composed mainly of quartz, calcite and dolomite. The presence of palygorskite in the samples collected in the current study, as it has been already discussed, is an extra evidence that the samples are originated from an arid environment and are characterized by alkaline pH (western Sahara) (Molinarioli 1996; Coz et al. 2009).

Moreover, Fig. 7 presents four diagnostic ratios to examine if the dust samples originate from the same desert region. The wet samples (S2, S3 and S4) with higher Ca/Al and Si/Al ratios have simultaneously lower Fe/Ca and K/Ca ratios. According to Avila et al. (1997) larger Si/Al and Ca/Al ratios can be ascribed to dust samples with a larger content of quartz, calcite and dolomite minerals coming from Moroccan Atlas (Blanco et al. 2003). Furthermore, the dust origin of the samples collected during the dust storms on 17/04/2005 and 05/03/2009 was also investigated by back-trajectory analysis (see also Figs. 4, 5). During the Sahara dust event that this sample (S2) was collected, the air mass came from Moroccan Atlas Mountains and the ratios (Ca/Si, Ca/Al and Si/Al) showed higher values (Fig. 7). In the contrary, the sample collected on the 5th of March 2009 (S4), came from the region of Egypt and Libya and had lower Ca/Si, Ca/Al and Si/Al ratios, but higher Fe/Ca and K/Ca ratios. These results in conjunction with the levels of illite, smectite, and kaolinite, affirm the assumption that the dust collected is originated from central Algeria or easterly African regions. It is likely that the ratio between two selected elements can be strongly affected by atmospheric processes such as turbulence, gravitational settling and other dry and wet atmospheric processes (Guerzoni et al. 1997). The above results show that the mineralogical differences between sources can be significant for the lower content minerals such as quartz, kaolinite, dolomite, and smectite.



**Fig. 7** Diagnostic ratios of the collected dust samples at Akrotiri station burst events

## 4 Conclusions

Sahara dust events were identified at the region of Akrotiri monitoring station in the period 2003–2013. The back-trajectory analysis of air masses and images from satellite sensors showed that there is a positive correlation between the air masses originated from North Africa and the elevated  $PM_{10}/PM_{2.5}$  concentrations in the region of Crete.

The highest probability for a Sahara dust event to occur is during spring and, the lowest during summer with a duration fluctuated from 2 to 78 h. Although the percentage of exceedances attributable to Sahara dust events was variable, in the years 2008 and 2010 more than 50% of the exceedances were attributed to Sahara dust bursts. The largest decrease in the average monthly PM concentrations after removing elevated PM concentrations due to Sahara dust events was observed during the 8-month period November to June (20.5%). The average yearly percentage of days with Sahara outbreaks in the region of west Crete was 9.3% for the period 2003–2013. This percentage is lower than the Sahara dust events in Cyprus and in Finokalia, where the percentages were 34 and 31%, respectively.

Mineralogical analysis showed that illite, quartz and calcite were the main components of mineral dust. Illite was the most abundant mineral identified in all samples. Gypsum was detected only in the dry sample, which means that in all wet” samples quantities of gypsum were dissolved in the rainwater. The lack of smectite suggests that air masses reaching Crete are originated from dust sources of Western Sahara and southern Morocco.

Furthermore, the elemental particle composition analysis showed that the values of Al/Si ratio were ranged from 0.31 to 0.39, in the dust samples originated from rainfall residues. This suggests that collected dust is originated from Africa deserts.

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