

## Abstract

The main aim of the present study is to describe the vertical structure of the intense Mediterranean dust outbreaks, based on the use of satellite and surface-based retrievals/measurements. Strong and extreme desert dust (DD) episodes are identified at  $1^\circ \times 1^\circ$  spatial resolution, over the period March 2000–February 2013, through the implementation of an updated objective and dynamic algorithm. According to the algorithm, strong DD episodes occurring at a specific place correspond to cases in which the daily aerosol optical depth at 550 nm ( $AOD_{550\text{nm}}$ ) exceeds or equals the long-term mean  $AOD_{550\text{nm}}$  (Mean) plus two standard deviations (SD) value being smaller than  $\text{Mean} + 4 \cdot \text{SD}$ . Extreme DD episodes correspond to cases in which the daily  $AOD_{550\text{nm}}$  value equals or exceeds  $\text{Mean} + 4 \cdot \text{SD}$ . For the identification of DD episodes additional optical properties (Angstrom exponent, fine fraction, effective radius and Aerosol Index) derived by the MODIS-Terra & Aqua (also AOD retrievals), OMI-Aura and EP-TOMS databases are used as inputs. According to the algorithm using MODIS-Terra data, over the period March 2000–February 2013, strong DD episodes occur more frequently (up to 9.9 episodes  $\text{yr}^{-1}$ ) over the western Mediterranean while the corresponding frequencies for the extreme ones are smaller (up to 3.3 episodes  $\text{yr}^{-1}$ , central Mediterranean Sea). In contrast to their frequency, dust episodes are more intense (AODs up to 4.1), over the central and eastern Mediterranean Sea, off the northern African coasts. Slightly lower frequencies and higher intensities are found when the satellite algorithm operates based on MODIS-Aqua retrievals, for the period 2003–2012. The performance of the satellite algorithm is assessed against surface-based daily data from 109 sun-photometric (AERONET) and 22  $PM_{10}$  stations. The agreement between AERONET and MODIS AOD is satisfactory ( $R = 0.505\text{--}0.75$ ) improving considerably when MODIS level 3 retrievals with higher sub-grid spatial representativeness and homogeneity are considered. Moreover, the evaluation analysis using other AERONET spectral optical and microphysical properties during the days of episodes as well as surface  $PM_{10}$  concentrations also provides strong support of the successful perfor-

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here, you have to state that these two products have been used ONLY over sea.

provide 2 or 3 digits

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et al., 2010; Huang et al., 2010) and Europe (e.g. Mona et al., 2006; Papayannis et al., 2008; Basart et al., 2012; Bègue et al., 2012; Pey et al., 2013), favored by the prevailing atmospheric circulation patterns, from planetary to synoptic scales. Due to their frequent transport in the Mediterranean, mineral dust particles, constitute the predominant aerosol type there (Barnaba and Gobbi, 2004; Basart et al., 2012), as shown by the good agreement, in spatial terms, between the geographical distributions of dust episodes' AOD (Gkikas et al., 2013) and average AOD conditions (Papadimas et al., 2008).

Dust particles play an important role for the shortwave (SW) and longwave (LW) radiation budget (e.g. Kaufman et al., 2002; Tegen et al., 2003; Heinold et al., 2008) and climate (IPCC, 2013). They affect atmospheric heating/cooling rates (e.g. Mallet et al., 2009) while they can also result in a modification of atmospheric dynamics and large atmospheric circulations like monsoons (e.g. Lau et al., 2006; Bollandina et al., 2011), cloud properties and precipitation (e.g. Huang et al., 2006; Solmon et al., 2008). Moreover, it has been shown that the consideration of their radiative impacts in numerical simulations can improve the forecasting accuracy of weather models (Pérez et al., 2006). Dust particles also affect air quality in urban areas (Basart et al., 2012) causing adverse health effects (Díaz et al., 2012; Karanasiou et al., 2012; Pérez García-Pando et al., 2014). All these consequences of dust aerosol are relevant and maximize under maximum dust loads, namely dust episodes, highlighting thus the significance of analyzing the spatial and temporal characteristics of such events. To this aim, many studies have been carried out using either surface (e.g. Cachorro et al., 2006) or satellite (e.g. Moulin et al., 1998) observations, as well as modelling techniques (e.g. Heinold et al., 2007) focusing on the broader Mediterranean area. These studies have been done either for specific cases (e.g. Kubilay et al., 2003; Balis et al., 2006) or for extended periods at specific locations (e.g. Meloni et al., 2007; Toledano et al., 2007a; Gobbi et al., 2013; Mona et al., 2014). Recently, Gkikas et al. (2013) developed an objective and dynamic algorithm relying on satellite retrievals, which enabled an overall view of dust episodes over the entire Mediterranean and the characterization of their regime.

Therefore, several

mechanisms

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Extensive research has been also carried out on the causes of Mediterranean dust outbreaks. Different mechanisms and processes of transport, apart from dust emissions in source areas, have been proposed as controlling factors. Moulin et al. (1997) showed that the exported dust loads from Northern Africa towards the Atlantic Ocean and the Mediterranean are controlled by the phase of the North Atlantic Oscillation (NAO). Other studies, focused on the description of atmospheric circulation characteristics favoring the occurrence of desert dust outbreaks over the central (Barkan et al., 2005; Meloni et al., 2008) or western (Querol et al., 1998; Rodriguez et al., 2001; Salvador et al., 2014) Mediterranean, but on a synoptic scale. An objective classification, based on multivariate statistical methods, of the atmospheric circulation patterns related to dust intrusions over the Mediterranean, has been presented by Gkikas et al. (2014) and Varga et al. (2014). The concentration of dust aerosols in the Mediterranean is characterized by strong spatial and temporal variability, associated with the seasonal variability of cyclones dominating or affecting the broader Mediterranean basin (Trigo et al., 2002). According to Moulin et al. (1998), dust AOD levels are higher in spring and summer compared to the wet seasons of the year. Moreover, dust intrusions are mainly recorded over the eastern Mediterranean in spring and winter, over the western parts in summer and over the central ones in autumn (Gkikas et al., 2013):

5 new paragraph

Dust transport over the Mediterranean is characterized by a multi-layered structure (Hamonou et al., 1999) in contrast to the Atlantic Ocean, which is well confined to the Saharan Air Layer (SAL, Karyampudi et al., 1999). The vertical distribution of dust particles into the troposphere and the profile of their physical and optical properties at different altitudes are controlling their impacts on atmospheric dynamics (Zhang et al., 2013). In order to describe the geometrical features of dust transport, many researchers have used ground lidar measurements, model simulations (Alpert et al., 2004; Kishcha et al., 2005) or they have relied on a synergistic use of satellite observations and ground lidar profiles (Berthier et al., 2006). The vertical extension of the Saharan dust intrusions over Europe, during the period 2000–2002, was the subject

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(Papayannis et al., 2008)

and Papayannis et al., 2008

This part is not clear. Please rephrase

⊗ This is false and is contradictive to the long-term measurements from EARLINET data base (Mona et al., 2012; Papayannis et al., 2008). The authors should rephrase and correct this sentence, explaining that their findings mostly concern the south-eastern Mediterranean region and not the eastern Mediterranean

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of a comprehensive study by Papayannis et al. (2008), who used lidar measurements from the EARLINET (European Aerosol Research Lidar Network, Bösenberg et al., 2003). Over the Mediterranean stations, the mean base, top and thickness of dust layers was found to vary from 1356 to 2980 m, 3600 to 5900 m and 726 to 3340 m, respectively. According to the obtained results, tracers of dust particles can be detected up to 10 km, as also reported by Gobbi et al. (2000), who studied a Saharan dust event in Crete (south Greece) during spring of 1999.

Several similar studies have been also performed for specific Mediterranean locations based on EARLINET lidar measurements. For example, Mona et al. (2006) analyzed the vertical structure of 112 Saharan intrusions that occurred over Potenza (Italy), from May 2000 to April 2003. The authors found that these outbreaks are confined between 1.8 and 9 km while their mass center is located at 3.5 km a.s.l. A similar analysis for Athens and Thessaloniki over the period 2000–2002, was conducted by Papayannis et al. (2005) who demonstrated that dust layers are recorded mainly between 2 and 5 km while their thicknesses vary from 200 to 3000 m. The geometrical characteristics of dust layers over Athens, during the period 2004–2006, have been also presented by Papayannis et al. (2009), who pointed out that the center of mass of dust layers is located at 2900 m being in a very good agreement with Kalivitis et al. (2007) findings (around 3 km) for the eastern Mediterranean. Additionally, the authors reported that the dust layers mainly extend from 1600 to 5800 m while mineral particles can be detected, at very low concentrations, up to 8 km a.s.l.. Gobbi et al. (2013) found that dust plumes, over Rome, mainly extend from 0 to 6 km while their center of mass is located at around 3 km. In the southern parts of Italy (Potenza), dust layers' base is found between 2 and 3 km, their geometrical height extends from 2.5 to 4 km while tracers of dust particles can be detected up to 10 km, based on a dataset of 310 dust events analyzed by Mona et al. (2014). Finally, Pisani et al. (2011) stated that the mean base and top of dust layers is found at 1500 and 4600 m a.s.l., respectively, while their mean thickness is equal to 3100 m, based on a statistical analysis of 45 desert dust episodes observed over Naples (Italy), from May 2000 to August 2003.

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Please be consistent : either you use "km" or "m"

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tail against surface measurements provided by AERONET or PM<sub>10</sub> stations, located within the study region (Sect. 4.2). Additionally, useful information about various optical and physical properties under intense dust episodes conditions is also derived from the evaluation analysis. For the identified DD episodes, collocated CALIOP-CALIPSO vertical feature mask and total backscatter coefficient retrievals are used in order to describe the annual and seasonal variability of dust outbreaks' vertical extension over the Mediterranean (Sect. 4.3). Finally, the summary and conclusions are drawn in Sect. 5.

## 2 Satellite and surface-based data

The different types of satellite retrievals that have been used as <sup>the</sup> inputs to the objective and dynamic satellite algorithm are described below, namely MODIS (Sect. 2.1.1), EP-TOMS and OMI-Aura (Sect. 2.1.2) databases. Also, CALIOP-CALIPSO vertically resolved satellite data, coincident with the identified desert dust outbreaks by the satellite algorithm, are described in Sect. 2.1.3. Finally, surface-based sun-photometric AERONET retrievals and PM<sub>10</sub> concentrations, both used to assess the performance of the satellite algorithm, are described in Sect. 2.2.1 and 2.2.2, respectively.

### 2.1 Satellite data

#### 2.1.1 MODIS

MODerate resolution Imaging Spectroradiometer (MODIS) onboard the Terra and Aqua satellites – with daytime local equator crossing time at 10:30 and 13:30, respectively, and 2330 km viewing swath – acquires measurements at 36 spectral bands between 0.415 and 14.235 μm with spatial resolution of 250, 500 and 1000 m. Observations from Terra and Aqua are made continuously since February 2000 and July 2002, respectively, and are available from the LAADS website (<ftp://ladsweb.nascom.nasa.gov/>). Aerosol optical properties are retrieved through the Dark Target (DT) algorithm (see e.g. Kaufman et al., 1997, 2001; Tanré et al., 1997; Levy et al., 2003; Remer et al.,

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2005) where different assumptions are considered depending on the underlying surface type (land or ocean). Several evaluation studies (e.g. Remer et al., 2008; Papadimas et al., 2009; Levy et al., 2010; Nabat et al., 2013) have shown that aerosol optical depth (AOD) can be retrieved satisfactorily by MODIS, nevertheless its performance is better over sea (uncertainty equal to  $\pm 0.03 \pm 0.05 \times \text{AOD}$ , Remer et al., 2002) than over land ( $\pm 0.05 \pm 0.15 \times \text{AOD}$ , Levy et al., 2010).

The following daily MODIS-Terra and MODIS-Aqua Collection 051 (C051) level 3 satellite data (MOD08\_D3 and MYD08\_D3 files) provided at  $1^\circ \times 1^\circ$  latitude-longitude spatial resolution are used: (i)  $\text{AOD}_{550 \text{ nm}}$ , (ii) Ångström exponent over land ( $\alpha_{470-660 \text{ nm}}$ ), (iii) Ångström exponent over ocean ( $\alpha_{550-865 \text{ nm}}$ ), (iv) fine-mode fraction (FF) of AOD over land and ocean and (v) Effective radius over ocean ( $r_{\text{eff}}$ ). Similar data have been used by Gkikas et al. (2013), however, in the present study we have improved data quality by using the quality assurance-weighted (QA) data only ([http://modis-atmos.gsfc.nasa.gov/\\_docs/QA\\_Plan\\_2007\\_04\\_12.pdf](http://modis-atmos.gsfc.nasa.gov/_docs/QA_Plan_2007_04_12.pdf)). Moreover, the day cloud fraction as well as the number of level 2 counts, which are both relevant to the performance of the satellite algorithm, are also used in this study. The time series of daily MODIS aerosol data cover the 13 yr period March 2000–February 2013 (Terra) and the 10 yr period January 2003–December 2012 (Aqua).

### 2.1.2 EP/TOMS and OMI-Aura

The selected retrievals from MODIS provide information about particles' load and size, which are both necessary to identify dust episodes. However, since dust is not the only coarse aerosol, for example sea-salt can be so as well, another optical property indicative of particle absorption efficiency is also required by the algorithm. To address this issue, the Absorption Aerosol Index (AI) daily data were also used, derived from measurements taken by the Total Ozone Mapping Spectrometer (TOMS) onboard the NASA's Earth-Probe satellite and the Ozone Monitoring Instrument (OMI) instrument onboard the NASA's Aura satellite. AI is the primary TOMS aerosol product (Herman et al., 1997) based on a spectral contrast method in a UV region (331–

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*(AOD data)*  
*(AE and FF data)*

*(\*) Although the authors state that they do not use AE and FF data over land, here they state that they are using these data! This is contradictory and not acceptable.*

360 nm) where ozone absorption is very small and can be used for the distinction between scattering (e.g. sea-salt) and absorbing (e.g. desert dust, smoke, volcanic ash) aerosols. The retrieval algorithm (fully described by Torres et al., 1998, 2002, 2005) takes advantage of the low surface albedo in the UV spectrum range, even in arid and semi-arid areas, making thus possible the estimation of the AOD over highly reflecting desert surfaces, where the major dust sources are located. Since the late 70's, the TOMS sensor onboard Nimbus-7 (1978–1993) and Earth Probe (1996–2005) has been providing global aerosol measurements. With the deployment of the EOS-Aura OMI (Ozone Monitoring Instrument) in mid-2004 (Torres et al., 2007) the near UV aerosol record continues to be extended into the foreseeable future. OMI is a hyperspectral sensor, covering the 270–500 nm range, launched onboard the EOS-Aura satellite on 15 July 2004 (13:38 equator crossing time, ascending mode) providing almost daily global coverage thanks to its wide viewing swath (2600 km with 13 km × 24 km nadir resolution). Apart from AI measurements, OMI aerosol products include also the total and absorption AOD and the single scattering albedo at 388 and 500 nm (Torres et al., 2007). Both EP-TOMS and OMI-Aura retrievals are available via the Mirador ftp server (<http://mirador.gsfc.nasa.gov/>) of the Goddard Earth Sciences Data and Information Services Center (GES DISC).

### 2.1.3 CALIOP-CALIPSO

The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard the NASA's satellite CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations), launched in April 2006; provides vertical resolved aerosol and cloud observations (Winker et al., 2009) since June 2006. CALIPSO is flying in the A-Train constellation (Stephens et al., 2002; <http://atrain.nasa.gov/>) in a sun-synchronous polar orbit at 705 km over the surface, with a 16-day repeat cycle, crossing the equatorial plane at about 13:30 LST (Winker et al., 2009). CALIOP is an active sensor measuring the backscatter signal at 532 and 1064 nm as well as the polarization at 532 nm (Winker et al., 2009). These level 1 retrievals are further processed (calibra-

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⊗ This is misleading not always the volcanic ash is absorbing. It is mainly scattering (containing sulfate aerosols).

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in more than 1000 locations of the planet (<http://aeronet.gsfc.nasa.gov>). The solar irradiances received by the photometer are inverted to columnar aerosol optical and microphysical properties through the implementation of retrieval algorithms (e.g. Dubovik and King, 2000; O'Neill et al., 2003). The followed standardized methods concerning instrument maintenance, calibration, cloud screening and data processing allow aerosol monitoring and comparison between different study periods and areas (Smirnov et al., 2000). From the global AERONET stations, 109 are located within the geographical limits of our study region. For each station, the daily averages of cloud-screened and quality assured data (Level 2.0) of direct sun and almucantar retrievals are used for: (i) AOD at 7 wavelengths from 340 to 1020 nm, (ii) size distribution retrieved for 22 logarithmically equidistant discrete points ( $r_i$ ) in the range of sizes  $0.05 \mu\text{m} \leq r \leq 15 \mu\text{m}$ , (iii) Ångström exponent between 440 and 870 nm ( $\alpha_{440-870 \text{ nm}}$ ), (iv) total effective radius ( $r_{\text{eff}}$ ), and (v) single scattering albedo (SSA) and asymmetry parameter ( $g_{\text{aer}}$ ) both retrieved at 440, 675, 870 and 1020 nm. The uncertainty in the estimation of AOD depends on technical (e.g. calibration method) factors and inversion assumptions, both described in detail in Holben et al. (1998). Moreover, the accuracy of the retrieved AOD by the CIMEL radiometer is spectrally dependent, being higher ( $< \pm 0.01$ ) for wavelengths longer than 440 nm and lower ( $< \pm 0.02$ ) for UV wavelengths (Eck et al., 1999). It should be also noted that the AERONET Level 2.0 inversion products (e.g. SSA) are provided when AOD at 440 nm is higher than 0.4 ensuring the minimization of the inversion uncertainties, which are also determined by other factors (e.g. scattering angle, particles' sphericity) as stated in detail by Dubovik et al. (2000).

**2.2.2 PM<sub>10</sub>**

Daily total and dust surface PM<sub>10</sub> concentrations, over the period 2001–2011 from 22 regional background and suburban background sites were used in this study. The monitoring sites are distributed as follows: 10 are located in Spain; 2 are in southern France; 5 are in Italy; 3 are in Greece; 1 is in southern Bulgaria and 1 is in Cyprus. PM<sub>10</sub> concentrations were obtained in most cases from gravimetric determinations on





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tions applied to the satellite algorithm inputs (e.g. implementation of QA MODIS retrievals as discussed in Sect. 2.1.1). The decreasing south-to-north gradient of intense DD episodes' frequency, which is also in agreement with previous studies based on ground PM measurements (Querol et al., 2009b; Pey et al., 2013), model simulations (Papayannis et al., 2008) and AERONET AOD retrievals (Basart et al., 2009), can be attributed to the increasing distance from the major dust sources and to the higher precipitation amounts at the northern parts of the basin (e.g. Marriotti et al., 2002; Mehta and Yang, 2008).

The maximum frequencies (9.9 episodes $\text{yr}^{-1}$ ) of strong DD episodes are observed in the western parts of the study region, for both periods and datasets, while the corresponding values for the extreme ones (3.3 episodes $\text{yr}^{-1}$ ) are observed over the central Mediterranean Sea for MODIS-Terra (March 2000–February 2013). In general, there is similar spatial variability between Terra and Aqua, though slightly lower maximum frequencies are found for Aqua. Although dust episodes occur rarely across the northern parts of the study region (< 1 and 0.5 episodes $\text{yr}^{-1}$  for strong and extreme episodes), their occurrence proves that dust particles can be transported far away from their sources, up to the central (e.g. Klein et al., 2010) or even northern (e.g. Bègue et al., 2012) European areas under favorable meteorological conditions. A noticeable difference between the two study periods and platforms is that relatively high frequencies of extreme DD episodes are recorded in more northern latitudes in the Mediterranean Sea, i.e. up to 43° N, according to MODIS-Terra over March 2000–February 2013, while they are restricted south of 40° N parallel for MODIS-Aqua during 2003–2012. In order to investigate this difference in detail we have also applied the satellite algorithm, over the period 2003–2012, i.e. that of Aqua, using MODIS-Terra retrievals as inputs. Through this analysis (results not shown here), it is evident that there is a very good agreement between the satellite algorithm's outputs, for the periods March 2000–February 2013 and 2003–2012, revealing a constant dust episodes' regime. Therefore, the discrepancy appeared between MODIS-Terra and MODIS-Aqua spatial distributions in Fig. 2, is attributed to the diurnal variation of factors regulat-

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Papayannis et al., 2014

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while the issue of possible cloud contamination is also considered. However, since the obtained results revealed a very similar performance of the algorithm for both periods and platforms, only the results for the period March 2000–February 2013 are given here.

5 In 46 out of 109 AERONET stations, depicted with yellow triangles in Fig. 4, <sup>we</sup> have been found at least one strong or extreme dust episode, for which coincident satellite and ground measurements are available. For the specific AERONET stations and episode days, the mean values of the selected AERONET aerosol optical properties have been calculated separately for strong, extreme and all (both strong and extreme) 10 DD episodes identified by the satellite algorithm. Subsequently, these values were compared to the corresponding ones calculated from all the available retrievals (climatological conditions, clim) collected from the 109 Mediterranean AERONET stations, during the period March 2000–February 2013, aiming at highlighting the effect of episodes on these optical properties. Additionally, in 7 AERONET stations (cyan circles in Fig. 4) the 15 intense DD episodes have been identified from ground and the corresponding results are compared with the satellite algorithm outputs (Sect. “Intercomparison of surface-based and satellite algorithms used for the identification of the desert dust episodes”). Finally, the performance of the algorithm is also tested against surface PM<sub>10</sub> measurements from 22 stations (Sect. 4.2.2).

#### 20 4.2.1 AERONET

##### Aerosol optical depth

During the period March 2000–February 2013, 346 pixel level intense DD episodes have been identified by the satellite-based algorithm, in which coincident MODIS-Terra and AERONET retrievals are available. It should be noted that AERONET AOD<sub>550 nm</sub> values have been calculated from available AERONET AOD<sub>870 nm</sub> and Ångström exponent data ( $\alpha_{440-870 \text{ nm}}$ ) by applying the Ångström equation (Ångström, 1929) to match the MODIS AOD<sub>550 nm</sub>. For these intense DD episodes, the comparison between the

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The accuracy of the DD episodes identification method was further evaluated by also using other AERONET aerosol optical properties than AOD, namely the Angström exponent ( $\alpha$ ) and the effective radius ( $r_{\text{eff}}$ ), able to provide information about particles' size. These two parameters, especially the first one, have been widely used by many researchers in order to identify or discriminate fine and coarse particles (e.g. O'Neill et al., 2003; Fotiadi et al., 2006; Toledano et al., 2007b; Gobbi et al., 2007; Basart et al., 2009; Prats et al., 2011). For this reason, we have produced the boxplots of  $\alpha$  (Fig. 8-i) and  $r_{\text{eff}}$  (Fig. 8-ii) values, derived by AERONET sun photometers, under climatological and intense dust episode conditions (strong, extreme and all). The appropriateness of our methodology and algorithm is confirmed by the drastic reduction of  $\alpha$  (Fig. 8-i) and increase of  $r_{\text{eff}}$  (Fig. 8-ii) values when dust outbreaks occur. Namely, the mean  $\alpha$  value decreases by a factor of 4.8 while the  $r_{\text{eff}}$  value increases by 2.5 times, under extreme DD episodes in the Mediterranean.

According to the boxplot statistics, when all available AERONET retrievals are considered (orange boxplot),  $\alpha$  ranges from  $-0.08$  to  $2.51$  with mean and median values equal to  $1.29$  and  $1.38$ , respectively. Furthermore, the majority ( $> 75\%$ ) of  $\alpha$  values is higher than  $1.04$  indicating the strong presence of fine particles in the study domain. On the contrary, during intense dust episodes the statistics are totally modified. For all DD episodes (green boxplots), the mean and median values are decreased down to  $0.43$  and  $0.27$ , respectively, while most of  $\alpha$  values ( $> 75\%$ ) are lower than  $0.54$ . Similar results are found for the strong DD episodes (cyan boxplots) while for the extreme cases (red boxplots) the corresponding statistical values are equal to  $0.27$ ,  $0.20$  and  $0.36$ , respectively. Similar findings were reported by Tafuro et al. (2006) who calculated a low average  $\alpha$  value equal to  $0.2 \pm 0.1$  during dusty days ( $\text{AOD}_{440\text{nm}} > 0.6$ ) from 5 AERONET stations located in the central Mediterranean, related with transported mineral particles from the northern African deserts (Pace et al., 2006). Basart et al. (2009) analyzed AERONET retrievals provided at fine temporal resolution (15 min) from 39

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In Fig. 10, <sup>we present</sup> ~~are presented~~ the overall scatterplots between satellite and ground AODs when intense DD episodes have been identified based on the ground (left column) and the satellite (right column) algorithm. Colors in Fig. 10i-a, ii-a, iii-a represent the associated MODIS-Terra Ångström exponent, effective radius and day cloud fraction (CFD) retrievals, respectively. In Fig. 10i-b and ii-b colors represent the AERONET Ångström exponent and effective radius, respectively, while in Fig. 10iii-b represent the day cloud fraction observations derived by MODIS-Terra. Through this approach it is feasible to evaluate furthermore the performance of the satellite algorithm, specify its drawbacks and check the validity of the defined thresholds (green boxes in Fig. 1).

It is apparent that the agreement between MODIS-Terra and AERONET AODs is better when DD episodes are identified from the ground, as shown by the increased correlation coefficients (from 0.52 to 0.7), increased slopes (from 0.6 to 0.9–1.0) and decreased biases (from 0.16 to –0.03). In particular, when DD episodes are identified from space, the MODIS-Terra AOD retrievals are overestimated (bias = 0.163) with regards to AERONET, particularly at low AOD values (< 0.5). In both cases, the highest overestimations are associated with cloud fractions higher than 0.7 due to the possible contamination of the satellite AODs by clouds (Fig. 10iii-a, iii-b). Given that DD episodes' identification based on AERONET retrievals is more efficient, we have used these outputs in order to check the validity of the defined thresholds in the satellite algorithm (green boxes in Fig. 1). For each aerosol optical property, it has been calculated the percentage of intense DD episodes for which the corresponding satellite observations are below or above the defined thresholds, depending on the parameter. The results given in Table 2 are satisfactory, since the percentages range from 87 to 99 %, and confirm the validity of the defined thresholds.

The scatterplots in Fig. 10i-b and ii-b also reveal some weaknesses of the satellite-based algorithm. More specifically, it is found that for few DD episodes identified by the satellite algorithm the corresponding AERONET Ångström exponent and effective radius values are higher than 1 and smaller than 0.4, respectively. These values indicate a predominance of fine particles instead of coarse ones as it would be expected

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formed a similar analysis but using data for a shorter period (2000–2007) than ours. It must be noted that the correlation coefficients are affected by outliers, because of the limited number of DD episodes in each station, highlighting the sensitiveness of the intercomparison. Such outliers can be expected when satellite-based columnar AODs and surface-based PM<sub>10</sub> data are compared, since satellite AODs are representative for the whole atmospheric column in contrast to in-situ PM measurements which are more representative for the planetary boundary layer affected also by local factors. Therefore, the vertical distribution of desert dust load, as it will be presented in the next section, can determine the level of agreement between satellite AODs and surface PM concentrations. Another influencing factor can be cloud contamination of MODIS AOD.

The identification method by the satellite algorithm can be considered as correct when dust PM<sub>10</sub> concentrations are higher than zero. According to this, the ratio between the number of non-zero dust PM observations and the number of DD episodes (coincident satellite-derived DD episodes and total PM<sub>10</sub> measurements) for each station is defined as success score. The calculated success scores (Fig. 11-iii) vary from 68 % (Monagrega, northeastern Spain, 28 episodes) to 97 % (Bocadifalco, Sicily, 33 episodes) confirming the appropriateness of the DD episodes' identification. In the majority of stations, the contribution of dust particles to the total burden (Fig. 11-iv) is above 50 %, ranging from 44 % (Zarra, Spain) to 86.8 % (Ayia Marina, Cyprus). In order to complete our analysis we have also calculated the mean (Fig. 11-v) and the median (Fig. 11-vi) dust PM<sub>10</sub> concentrations for the identified intense DD episodes in each station. The mean PM<sub>10</sub> concentrations mainly vary between 20 and 50 μg m<sup>-3</sup>, being higher in the southern stations, as expected. The minimum mean value (17 μg m<sup>-3</sup>) was recorded in Censt (Sardinia) and the maximum one (223 μg m<sup>-3</sup>) in Ayia Marina (Cyprus). Our values are much higher than the corresponding ones in Querol et al. (2009b), who obtained that the mean levels of mineral matter in PM<sub>10</sub> during dusty days range from 8 to 23 μg m<sup>-3</sup> based on ground concentrations derived by 21 Mediterranean stations. These differences are reasonable since here only intense desert dust outbreaks associated with high aerosol optical depths are considered. Finally, the me-

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dian  $PM_{10}$  concentrations are lower compared to the average ones, indicating that outliers (cases with extremely high AOD or  $PM_{10}$ ) can alter the results, attributed to the fact that both parameters' (AOD and  $PM_{10}$ ) distributions are not Gaussians. For this reason the highest differences are found in Finokalia (Crete) and Ayia Marina (Cyprus), where the maximum daily  $PM_{10}$  concentrations, equal to 690 and 1291  $\mu g m^{-3}$ , respectively, were recorded during an intense dust outbreak affected the eastern Mediterranean on 24 and 25 February 2006.

**4.3 Vertical structure of the Mediterranean desert dust outbreaks**

The ability of the developed satellite algorithm to detect intense dust episodes has been proved adequate through the evaluation analysis against AERONET retrievals and  $PM_{10}$  concentrations. Nevertheless, its main limitation is that it uses satellite retrievals representative for the whole atmospheric column prohibiting thus the description of the vertical structure of these dust outbreaks. In order to address this issue, the CALIOP-CALIPSO retrievals are used as a complementary tool to the satellite algorithm's outputs. First, for the identified dust episodes by the satellite algorithm, the spatially and temporally collocated vertically resolved CALIOP lidar observations are selected. For these cases and for each  $1^\circ \times 1^\circ$  grid cell, we have divided the lower troposphere, up to 8 km, in 16 layers of 500 m height. In this way, 14 400 boxes of  $1^\circ \times 1^\circ$  surface area and 500 m height have been produced. Then, for each one of them, we have calculated the overall number of dust and polluted dust observations (hereafter named as dust) according to the aerosol subtyping scheme of the CALIOP Vertical Feature Mask (VFM). Note that dust and polluted dust were chosen because in previous studies (Mielonen et al., 2009) they were shown to be the best two defined aerosol types among the other ones classified by the CALIOP VFM. Nevertheless, in case of polluted dust, Burton et al. (2013) reported that dust particles can be mixed with marine aerosols instead of smoke or pollution as assumed by the VFM retrieval algorithm. In our study, more than 95 % of the aerosol type records were pure dust, for the collocated cases between the satellite algorithm and CALIPSO observations. In addition, in

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the majority of the defined boxes, the percentage of dust from the overall observations is higher than 70 %, confirming furthermore the validity of the algorithm DD episodes' identification procedure. This is an excellent proof of the successful identification of DD episodes by the satellite algorithm, since CALIOP-CALIPSO is an independent and vertically resolved platform and database. Thereby, CALIOP vertical observations were subsequently used to examine the vertical structure of dust outbreaks.

In order to analyze the intensity of desert dust outbreaks at different altitudes in the troposphere, the CALIOP data of total backscatter coefficient at 532 nm ( $\beta_{532\text{ nm}}$ ) have been also acquired. For each box, the average  $\beta_{532\text{ nm}}$  values have been calculated from all the available CALIOP measurements (day and night), for the identified intense dust episodes by the satellite algorithm. More specifically, the average  $\beta_{532\text{ nm}}$  values were calculated for the dust observations based on the CALIOP VFM associated with CAD scores ranging from  $-100$  to  $-20$ , as it has been proposed by Winker et al. (2013) for discriminating aerosol from clouds. The selection of  $\beta_{532\text{ nm}}$  values instead of extinction coefficients ensures that incorrect lidar ratio assumptions in the CALIOP retrieval algorithm do not affect our results. In the literature, it has been documented that the CALIOP lidar ratio is underestimated over the northern African deserts and the surrounding areas affected by Saharan dust particles, leading to an underestimation of the columnar AOD compared to MODIS and AERONET retrievals (Redemann et al., 2012; Schuster et al., 2012). Amiridis et al. (2013) stated that an increase of the lidar ratio from 40 to 58 sr, along with a series of post-corrections in the CALIOP retrievals and the implementation of several criteria concerning the cloud coverage and the spatial representativeness, can improve substantially the agreement between MODIS-Aqua/AERONET and CALIOP observations.

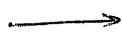
It should be noted that in the present work, we have analyzed all the available CALIOP overpasses ( $\sim 10\,000$ ) over the study region, during the period June 2006–February 2013. For brevity reasons, however, only the obtained results based on MODIS-Terra retrievals are presented here, since similar findings are drawn for MODIS-Aqua (June 2006–December 2012). Moreover, the analysis (results are not shown

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here) has been made separately for the identified strong and extreme DD episodes without revealing differences in the geometrical characteristics of dust outbreaks. Nevertheless, the  $\beta_{532\text{nm}}$  values are higher for the extreme DD episodes being consistent with the discrimination of dust episodes' intensity (in terms of AOD) which is applied to the satellite algorithm. In order to facilitate the visualization of our results, for each column ( $1^\circ \times 1^\circ$  spatial resolution) and latitudinal/longitudinal zone ( $1^\circ$ ), we have calculated the overall number of dust observations and the associated weighted averages of  $\beta_{532\text{nm}}$ , depending on the projection plane (latitudinal, longitudinal and columnar), according to dust observations in each box. For both parameters, the analysis has been made on an annual and seasonal basis and the corresponding results are discussed in Sect. 4.3.1 and 4.3.2, respectively.

## 4.3.1 Annual



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In Fig. 12, are presented the three dimensional structures of the CALIOP overall dust observations (Fig. 12-i) and the associated total backscatter coefficients at 532 nm (Fig. 12-ii), during intense dust episodes conditions, over the broader Mediterranean area, for the period June 2006–February 2013. From the latitudinal projection in Fig. 12-i, it is evident that dust particles are mainly detected between 0.5 and 6 km, and more rarely up to 8 km, between the parallels 32 and 38° N. The number of dust observations is increased at higher altitudes with increasing latitudes, up to 40° N, while the altitude range (thickness) where these records are detected is gradually reduced from 4 to 2 km. At northern latitudes, the CALIPSO dust records are drastically reduced and are mainly observed between 1 and 4 km. The ascending mode of the transported mineral particles over the Mediterranean is attributed to the prevailing low pressure systems, which mobilize and uplift dust particles from the source areas across the Sahara Desert and the Arabian Peninsula. Dust aerosols are transported over the planetary boundary layer (Hamonou et al., 1999) due to the upward movement of dry and turbid air masses (Dulac et al., 1992), while the prevailing synoptic conditions determine also the spatial

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and temporal characteristics of desert dust outbreaks over the Mediterranean (Gkikas et al., 2014).

In general, our results are in agreement with previous studies, based on lidar profiles, which have been made in several Mediterranean sites. More specifically, Papayannis et al. (2008) found that dust layers, over the EARLINET Mediterranean stations, extend from 0.5 to 10 km above mean sea level, their center of mass is located between 2.5 and 3.5 km and their thickness ranges from 2.1 to 3.3 km. Hamonou et al. (1999) reported that dust layers are mainly detected between 1.5 and 5 km based on lidar measurements in the northwestern and northeastern Mediterranean. According to di Sarra et al. (2001), who studied the Saharan dust intrusions in Lampedusa (central Mediterranean) for the period May–June 1999, dust particles can be detected up to 7–8 km, which is in line with our findings for the corresponding latitudinal zones (35–36° N). Balis (2012), analyzed 33 Raman/lidar profiles of Saharan dust intrusions over Thessaloniki (northern Greece), and found that the mean base and top of dust layers were equal to  $2.5 \pm 0.9$  and  $4.2 \pm 1.5$  km, respectively.

As to the variation of vertical extension with longitude (Fig. 12-i), it is revealed that the base height of dust layers is decreased towards the eastern parts of the study region. In the western Mediterranean, the mineral particles are mainly detected between 2 and 6 km while over the central and eastern Mediterranean the corresponding altitudes are equal to 0.5 and 6 km, respectively. It is well known, that dust is transported over the western Mediterranean mainly in summer (e.g. Moulin et al., 1998) favored by low pressure systems located over the northwestern Africa (Gkikas et al., 2014) and the enhanced thermal convection, uplifting effectively dust aerosols at high altitudes in the troposphere. Moreover, air masses carrying dust particles are "pushed" towards higher altitudes due to the existence of the Atlas Mountains Range. Therefore, the combination of strong convective processes over North Africa along with topography can explain the identification of dust aerosols at higher tropospheric levels over the western Mediterranean. It is the presence of mineral particles at high altitudes in western Mediterranean that can explain the poor-to-moderate agreement between

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PM<sub>10</sub> concentrations and MODIS AODs found in the Iberian Peninsula (Fig. 11-ii). On the contrary, air masses carrying African dust aerosols travel at lower altitudes over Africa and the central and eastern Mediterranean, because of absence of significant topographical objects on their route, as suggested by Pey et al. (2013).

5 Previous studies have shown that dust layers over the Mediterranean are characterized by a multilayered structure (e.g. Hamonou et al., 1999; Mona et al., 2006; Papayannis et al., 2008). This is also depicted in the longitudinal projection of Fig. 12-i, where several dust layers of different base/top altitudes and geometrical thicknesses are detected. In general, the base heights vary from 0.5 to 2 km, the top heights from  
10 4 to 6 km and the thicknesses from 1 to 4 km. The majority of common observations between the CALIOP profiles and the identified intense DD episodes by the satellite algorithm are recorded over the maritime parts of the study region (bottom map of Fig. 12-i). The maximum number of CALIOP dust observations (~ 19 000) is recorded along the Atlantic coasts of Morocco, but high numbers (about 10 000–15 000) are also  
15 found across the northern African coasts.

Apart from the CALIOP dust observations, we have also analyzed the associated  $\beta_{532\text{nm}}$  values at the defined altitude ranges in order to describe the variation of intensity of the desert dust episodes with height over the Mediterranean (Fig. 12-ii). The maximum backscatter coefficients (up to  $0.006\text{ km}^{-1}\text{ sr}^{-1}$ ) are observed below 2 km, being increased towards the southern edges (30–34° N) of the study region, where their source areas are found. This is explained by the fact that dust particles due to their coarse size and large mass, are efficiently deposited and for this reason they are recorded at higher concentrations near to the source areas and at low altitudes. Nevertheless, the decreasing intensity with height towards the north is not so evident. Thus,  
20 high  $\beta_{532\text{nm}}$  values ( $\sim 0.004\text{ km}^{-1}\text{ sr}^{-1}$ ) are observed between 2 and 4 km in the latitudinal zone extending from 35 to 42° N. Though, the uppermost altitudes where relatively high  $\beta_{532\text{nm}}$  values gradually decrease from 6 to 4 km, moving from South to North. Any differences in the latitudinal patterns of dust observations and backscatter values  
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(Fig. 12-i and ii) can be explained by the fact that  $\beta_{532\text{nm}}$  values take into account only the dust records and not the overall observations (all aerosol types).

The decrease of backscatter values at higher altitudes has been pointed out in previous studies where lidar profiles have been analyzed over specific Mediterranean locations (e.g. Mona et al., 2006; Papayannis et al., 2008). Nevertheless, it must be considered that in the aforementioned studies the lidar measurements are valid above the retrieved planetary boundary layer (Matthias et al., 2004) which varies depending on the location and the season (McGrath-Spangler et al., 2013). Despite the good agreement, as it concerns the vertical shape of the  $\beta_{532\text{nm}}$  curves, between our findings and the corresponding ones based on ground retrievals, in the present analysis the calculated backscatter coefficients are in general higher, which is reasonable since are considered only cases of intense desert dust outbreaks.

The longitudinal pattern of  $\beta_{532\text{nm}}$  profiles (Fig. 12-ii) is less distinct compared to the corresponding one resulted from the latitudinal projection. Relatively high  $\beta_{532\text{nm}}$  values ( $\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) are found between 1 and 5 km over the western Mediterranean, while over the central and eastern parts of the study region the desert dust outbreaks' intensity ( $\sim 0.006 \text{ km}^{-1} \text{ sr}^{-1}$ ) is higher below 1.5 km. Among the sub-regions, the backscatter coefficients are higher in the central and eastern Mediterranean, which is also depicted in the bottom map of Fig. 12-ii. It is reminded that higher intensities of dust episodes over the central and eastern Mediterranean have also been noticed based on MODIS retrievals (Fig. 3). From the obtained longitudinal projection, it is evident a patchy structure of the total backscatter coefficient profiles, especially in the central and eastern parts, indicating the existence of several dust layers of varying intensities at different altitudes into the atmosphere.

#### 4.3.2 Seasonal ~~variability~~ evolution? variability? please define

The vertical structure of Mediterranean desert dust outbreaks has also been analyzed separately for winter (DJF), spring (MAM), summer (JJA) and autumn (SON). The seasonal three dimensional representations of the CALIOP overall dust observations and

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The seasonal patterns of  $\beta_{532\text{nm}}$  latitudinal projections are different than those for the dust observations, while they also differ among the four seasons. The intensity of winter DD episodes is stronger (up to  $0.012\text{ km}^{-1}\text{ sr}^{-1}$ ) below 2 km and at the southern parts of the study region. According to the longitudinal and bottom map projections, these episodes take place over the central and eastern Mediterranean Sea but the number of grid cells with coincident CALIOP observations and DD episodes is limited. In spring, the highest  $\beta_{532\text{nm}}$  values (up to  $0.006\text{ km}^{-1}\text{ sr}^{-1}$ ) are recorded between the parallels 31 and  $35^\circ\text{N}$  and below 2 km, although, relatively high  $\beta_{532\text{nm}}$  values (up to  $0.004\text{ km}^{-1}\text{ sr}^{-1}$ ) are found up to 6 km (Fig. 13ii-b). Moving northwards, over the Mediterranean, dust layers are mainly confined between 2 and 4 km, associated with high  $\beta_{532\text{nm}}$  values (up to  $0.004\text{ km}^{-1}\text{ sr}^{-1}$ ) in the latitudinal zone extending from  $35$  to  $43^\circ\text{N}$ . The existence of these elevated dust layers, has been also confirmed by model simulations through specific (Papayannis et al., 2008) or averaged (Alpert et al., 2004) cross sections of dust concentrations in the central sector of the Mediterranean. This is in accordance with our longitudinal projection (Fig. 13ii-b), where  $\beta_{532\text{nm}}$  is high varying from 0.004 to  $0.008\text{ km}^{-1}\text{ sr}^{-1}$  at these altitude ranges.

In summer, the intensity of dust episodes is smoothly decreased at higher altitudes, where dust layers of considerable  $\beta_{532\text{nm}}$  values are also found. More specifically, the highest backscatter coefficients (up to  $0.008\text{ km}^{-1}\text{ sr}^{-1}$ ) are recorded near to the surface but also moderate values (up to  $0.006\text{ km}^{-1}\text{ sr}^{-1}$ ) are observed between 2 and 5 km, particularly over the southern parts of the study region (Fig. 13iii-b). Most of these intense DD episodes occur in the western Mediterranean, where the highest  $\beta_{532\text{nm}}$  values (up to  $0.005\text{ km}^{-1}\text{ sr}^{-1}$ ) are recorded between 2 and 5 km. Over the central and eastern Mediterranean, even higher  $\beta_{532\text{nm}}$  values are found (up to  $0.014\text{ km}^{-1}\text{ sr}^{-1}$ ) but at lower altitudes ( $< 1\text{ km}$ ). In autumn, the majority of the grid cells of coincident CALIOP profiles and DD episodes identified by the satellite algorithm are located between the parallels  $33$  and  $41^\circ\text{N}$ . In this latitudinal zone, CALIOP profiles are available over the interior parts of the Iberian Peninsula and over western and central parts of the Mediterranean Sea, near to the northern African coasts. According to the latitudi-

cerning the intense DD episodes' frequency (in terms of episodes  $\text{yr}^{-1}$ ) and intensity (in terms of AOD at 550 nm) are the following:

- 5 - Strong DD episodes occur more frequently (up to 9.9 episodes  $\text{yr}^{-1}$ ) in the western Mediterranean while the extreme ones occur more frequently (up to 3.3 episodes  $\text{yr}^{-1}$ ) over the central parts of the Mediterranean Sea, when the satellite algorithm operates with MODIS-Terra retrievals.
- Frequencies of occurrence of strong and extreme DD episodes are gradually reduced from south to north, while for the strong ones a west-east gradient is apparent.
- 10 - The intensity of strong and extreme DD episodes, in AOD terms, can reach up to 1.5 and 3-4, respectively, over the central and eastern parts of the Mediterranean Sea, near off the northern African coasts.
- Slightly lower frequencies and higher intensities are found for the period 2003-2012, when the satellite algorithm operates with MODIS-Aqua retrievals.
- 15 - The frequencies of occurrence and the intensities of DD episodes are slightly higher and lower, respectively, compared to the corresponding ones of the previous version of the satellite algorithm (Gkikas et al., 2013), applied for the period March 2000-February 2007, when MODIS-Terra non weighted QA retrievals were used as inputs.
- 20 Through a detailed evaluation of the satellite algorithm against surface measurements derived from 109 AERONET and 22  $\text{PM}_{10}$  stations, it is found that:

#### AERONET

- 25 - The correlation coefficient between MODIS and AERONET AODs is increased from 0.505 to 0.75 when level 3 grid cells with higher sub-grid spatial representativeness and homogeneity are considered.

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- Under intense dust episodes conditions, the spectral AERONET AOD median levels vary from 0.64 (340 nm) to 0.44 (1020 nm).
- According to the AERONET volume size distributions, it is evident the predominance of the coarse mode with a peak ( $\sim 0.25 \mu\text{m}^3 \mu\text{m}^{-2}$ ) for particles radii between 1.7 and 2.24  $\mu\text{m}$ , in case of intense DD episodes.
- The appropriateness of DD episodes' identification method applied to the satellite algorithm is confirmed since the majority (> 75 %) of AERONET  $\alpha_{440-870\text{nm}}$  and  $r_{\text{eff}}$  values are lower than 0.54 and higher than 0.55  $\mu\text{m}$ , respectively.
- The spectral variation of AERONET SSA and  $g_{\text{aer}}$  is found to be typical for desert dust aerosols. SSA values vary from 0.90 to 0.965 and increase (less absorption) with increasing wavelengths while  $g_{\text{aer}}$  varies from 0.704 (870 nm) to 0.742 (440 nm) revealing a reduced spectral variation.
- About 15 % of the pixel level intense DD episodes are misclassified by the satellite algorithm and these drawbacks are encountered in AERONET stations where the aerosol load is dominated either by fine particles or by complex aerosol types.

#### PM<sub>10</sub> and dust contribution

- The agreement between surface and satellite measurements is better over the central and eastern Mediterranean stations.
- On a station level, the percentage of the intense DD episodes, for which a dust contribution to PM<sub>10</sub> surface concentration has been recorded, varies from 68 % (Monagrega, northeastern Spain) to 97 % (Bocadifalco, Sicily).
- In the majority of stations, dust particles contribute more than 50 % of the total amount reaching up to 86.8 % (Ayia Marina, Cyprus).
- The mean PM<sub>10</sub> concentration levels mainly vary from 20 to 50  $\mu\text{g m}^{-3}$  reaching up to 223  $\mu\text{g m}^{-3}$  in Ayia Marina (Cyprus).

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