

# Mediterranean intense desert dust outbreaks and their vertical structure based on remote sensing data

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## 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 Mar. 2000 – Feb. 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 550nm ( $AOD_{550nm}$ ) exceeds or equals the long-term mean  $AOD_{550nm}$  ( $Mean$ ) plus two standard deviations ( $Std$ ), but being smaller than  $Mean+4*Std$ . Extreme DD episodes correspond to cases in which the daily  $AOD_{550nm}$  value equals or exceeds  $Mean+4*Std$ . For the identification of DD episodes, additional optical properties (Ångström 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 Mar. 2000 – Feb. 2013, strong DD episodes occur more frequently (up to 9.9 episodes  $yr^{-1}$ ) 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 consistency of the algorithm is successfully tested through the application of an alternative methodology for the determination of DD episodes, which produced similar features of the episodes' frequency and intensity, with just slightly higher frequencies and lower intensities. 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 ( $R=0.505\text{--}0.750$ ) improving considerably when MODIS level 3 retrievals with higher sub-grid spatial representativeness and homogeneity are considered. Through the comparison against  $PM_{10}$  concentrations, it is found that the presence of dust is justified in all ground stations with success scores ranging from 68 % to 97%. However, it is revealed a poor agreement between satellite and ground  $PM_{10}$  observations in the western parts of the Mediterranean attributed to the desert dust outbreaks' vertical extension and the high altitude of dust presence. The CALIOP vertical profiles of pure and polluted dust observations and the associated total backscatter coefficient at 532 nm ( $\beta_{532nm}$ ), indicate that dust particles are mainly detected between 0.5 and 6 km, though they can reach 8 km between the parallels 32° N and 38° N in warm seasons. An increased number of CALIOP dust records at higher altitudes is observed with increased latitude, northwards to 40° N, revealing an ascending mode of the dust transport. However, the overall intensity of DD episodes is maximum (up to  $0.006 \text{ km}^{-1} \text{ sr}^{-1}$ ) below 2 km and at the southern parts of the study region (30° N - 34° N). Additionally, the average thickness of dust layers gradually decreases from 4 to 2 km moving from South to North. In spring, dust layers of moderate-to-high  $\beta_{532nm}$  values ( $\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) are detected over the Mediterranean (35° N - 42° N), extending from 2 to 4 km. Over the western Mediterranean, dust layers are observed between 2 and 6 km, while their base height is decreased down to 0.5 km for increasing longitudes underlying the role of topography and thermal convection. The vertical profiles of CALIOP  $\beta_{532nm}$  confirm the multilayered structure of the Mediterranean desert dust outbreaks on both annual and seasonal bases, with several dust layers of variable geometrical characteristics and intensities. A detailed analysis of the vertical structure of specific DD episodes using CALIOP profiles reveals that the consideration of the dust vertical structure is necessary when attempting comparisons between columnar MODIS  $AOD$  retrievals and ground  $PM_{10}$  concentrations.

## 66 1. Introduction

67 The Mediterranean basin, due to its proximity to the major dust source arid areas of Northern  
68 Africa and Middle East (Middleton and Goudie, 2001; Prospero et al., 2002; Ginoux et al., 2012) is  
69 frequently affected by transported high dust loads referred to as episodes or events. The suspension and  
70 accumulation of mineral particles into the atmosphere over the Saharan and Arabian Peninsula's  
71 deserts are determined by various factors such as the enhanced turbulence, soil conditions (reduced  
72 vegetation cover and soil moisture), reduced precipitation amounts, latitudinal shift of the Intertropical  
73 Convergence Zone (ITCZ) as well as by small scale meteorological processes (e.g. haboobs). However,  
74 dust particles can be transported far away from their sources, mainly towards the Atlantic Ocean (e.g.  
75 Prospero and Lamb, 2003; Ben-Ami et al., 2010; Huang et al., 2010) and Europe (e.g. Mona et al.,  
76 2006; Mona et al., 2012; Papayannis et al., 2008; Basart et al., 2012; Bègue et al., 2012; Pey et al.,  
77 2013), favored by the prevailing atmospheric circulation patterns, from planetary to synoptic scales.  
78 Due to their frequent transport in the Mediterranean, mineral dust particles, constitute the predominant  
79 aerosol type there (Barnaba and Gobbi, 2004; Basart et al., 2012), as shown by the good agreement, in  
80 spatial terms, between the geographical distributions of dust episodes' *AOD* (Gkikas et al., 2013) and  
81 average *AOD* conditions (Papadimas et al., 2008).

82 Dust particles play an important role for the shortwave (SW) and longwave (LW) radiation budget  
83 (e.g. Kaufman et al., 2002; Tegen et al., 2003; Heinold et al., 2008; Sicard et al., 2014; Nabat et al.,  
84 2015) and climate (IPCC, 2013). They affect atmospheric heating/cooling rates (e.g. Mallet et al.,  
85 2009) while they can also result in a modification of atmospheric dynamics and large atmospheric  
86 circulations like monsoons (e.g. Lau et al., 2006; Bollasina et al., 2011), cloud properties and  
87 precipitation (e.g. Huang et al., 2006; Solomon et al., 2008). Moreover, it has been shown that the  
88 consideration of their radiative impacts in numerical simulations can improve the forecasting accuracy  
89 of weather models (Pérez et al., 2006). Dust particles also affect air quality in urban areas (Basart et al.,  
90 2012) causing adverse health effects (Díaz et al., 2012; Karanasiou et al., 2012; Pérez García-Pando et  
91 al., 2014). All these consequences of dust aerosol are relevant and maximize under maximum dust  
92 loads, namely dust episodes, highlighting thus the significance of analyzing the spatial and temporal  
93 characteristics of such events. To this aim, many studies have been carried out using either surface (e.g.  
94 Cachorro et al., 2006) or satellite (e.g. Moulin et al., 1998) observations, as well as modelling  
95 techniques (e.g. Heinold et al., 2007) focusing on the broader Mediterranean area. These studies have  
96 been done either for specific cases (e.g. Kubilay et al., 2003; Balis et al., 2006) or for extended periods  
97 at specific locations (e.g. Meloni et al., 2007; Toledano et al., 2007a; Gobbi et al., 2013; Mona et al.,

98 2014). Recently, Gkikas et al. (2013) developed an objective and dynamic algorithm relying on satellite  
99 retrievals, which enabled an overall view of dust episodes over the entire Mediterranean and the  
100 characterization of their regime (i.e., frequency of occurrence, intensity and duration).

101 Extensive research has also been carried out on the mechanisms of Mediterranean dust outbreaks.  
102 Therefore, several mechanisms and processes of transport, apart from dust emissions in source areas,  
103 have been proposed as controlling factors. Moulin et al. (1997) showed that the exported dust loads  
104 from Northern Africa towards the Atlantic Ocean and the Mediterranean are controlled by the phase of  
105 the North Atlantic Oscillation (NAO). Other studies, focused on the description of atmospheric  
106 circulation characteristics favoring the occurrence of desert dust outbreaks over the central (Barkan et  
107 al., 2005; Meloni et al., 2008) or western (Querol et al., 1998; Rodriguez et al., 2001; Salvador et al.,  
108 2014) Mediterranean, but on a synoptic scale. An objective classification, based on multivariate  
109 statistical methods, of the atmospheric circulation patterns related to dust intrusions over the  
110 Mediterranean, has been presented by Varga et al. (2014) and Gkikas et al. (2015).

111 The concentration of dust aerosols in the Mediterranean is characterized by strong spatial and  
112 temporal variability, associated with the seasonal variability of cyclones dominating or affecting the  
113 broader Mediterranean basin (Trigo et al., 2002). According to Moulin et al. (1998), dust *AOD* levels  
114 are higher in spring and summer compared to the wet seasons of the year. Moreover, dust intrusions are  
115 mainly recorded over the southeastern Mediterranean in spring and winter, over the western parts in  
116 summer and over the central ones in autumn (Gkikas et al., 2013).

117 Dust transport over the Mediterranean is characterized by a multi-layered structure (Hamonou et  
118 al., 1999; Papayannis et al., 2008) in contrast to the Atlantic Ocean, which is well confined to the  
119 Saharan Air Layer (SAL, Karyampudi et al., 1999). The vertical distribution of dust load into the  
120 troposphere as well as the profile of dust aerosols' optical properties at different altitudes, control the  
121 impacts on atmospheric dynamics induced by the mineral particles (Zhang et al., 2013). In order to  
122 describe the geometrical features of dust transport, many researchers have used ground lidar  
123 measurements, model simulations (Alpert et al., 2004; Kishcha et al. 2005) or they have relied on a  
124 synergistic use of satellite observations and ground lidar profiles (Berthier et al., 2006). The vertical  
125 extension of the Saharan dust intrusions over Europe, during the period 2000-2002, was the subject of a  
126 comprehensive study by Papayannis et al. (2008), who used lidar measurements from the EARLINET  
127 (European Aerosol Research Lidar Network, Bösenberg et al., 2003). Over the Mediterranean stations,  
128 the mean base, top and thickness of dust layers was found to vary from 1356 to 2980 m, 3600 to 5900

129 m and 726 to 3340 m, respectively. According to the obtained results, tracers of dust particles can be  
130 detected up to 10 km, as also reported by Gobbi et al. (2000), who studied a Saharan dust event in  
131 Crete (South Greece) during spring of 1999.

132 Several similar studies have been also performed for specific Mediterranean locations based on  
133 EARLINET lidar measurements. For example, Mona et al. (2006) analyzed the vertical structure of 112  
134 Saharan intrusions that occurred over Potenza (Italy), from May 2000 to April 2003. The authors found  
135 that these outbreaks are confined between 1.8 and 9 km while their mass center is located at 3.5 km  
136 above sea level (a.s.l.). A similar analysis for Athens and Thessaloniki over the period 2000-2002, was  
137 conducted by Papayannis et al. (2005) who demonstrated that dust layers are recorded mainly between  
138 2 and 5 km while their thicknesses vary from 0.2 to 3 km. The geometrical characteristics of dust layers  
139 over Athens, during the period 2004 – 2006, have been also presented by Papayannis et al. (2009), who  
140 pointed out that the center of mass of dust layers is located at 2.9 km being in a very good agreement  
141 with Kalivitis et al. (2007) findings (around 3 km) for the eastern Mediterranean. Additionally, the  
142 authors reported that the dust layers mainly extend from 1.6 to 5.8 km while mineral particles can be  
143 detected, at very low concentrations, up to 8 km a.s.l.. Gobbi et al. (2013) found that dust plumes over  
144 Rome mainly extend from 0 to 6 km while their center of mass is located at around 3 km. In the  
145 southern parts of Italy (Potenza), dust layers' base is found between 2 and 3 km, their geometrical  
146 height extends from 2.5 to 4 km while tracers of dust particles can be detected up to 10 km, based on a  
147 dataset of 310 dust events analyzed by Mona et al. (2014). Finally, Pisani et al. (2011) stated that the  
148 mean base and top of dust layers is found at 1.5 km and 4.6 km a.s.l., respectively, while their mean  
149 thickness is equal to 3.1 km, based on a statistical analysis of 45 desert dust episodes observed over  
150 Naples (Italy), from May 2000 to August 2003.

151 Surface-based lidar measurements like those used in the aforementioned studies provide useful  
152 information about the geometrical and optical properties of dust layers, but they are representative only  
153 for specific locations. Yet, a more complete knowledge about the vertical structure of dust outbreaks is  
154 necessary in order to adequately understand and determine their possible effects. The geographical  
155 limitation imposed by the use of surface-based lidar observations can be overcome by utilizing accurate  
156 satellite retrievals, as a complementary tool, which provide extended spatial coverage. Since 2006,  
157 vertical resolved observations of aerosols and clouds from space were made possible thanks to the  
158 CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) lidar flying onboard the CALIPSO  
159 (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite (Winker et al., 2009).  
160 Based on CALIOP observations, Liu et al. (2008) analyzed the global vertical distribution of aerosols

161 for one year, while other studies focused on the vertical structure of dust outflows towards the Atlantic  
162 Ocean (e.g. Ben-Ami et al., 2009; Adams et al., 2012; Tsamalis et al., 2013) and the Pacific Ocean (e.g.  
163 Eguchi et al., 2009; Hara et al., 2009). On the contrary, over the broader Mediterranean area, only a  
164 small number of studies has been made aiming at describing the vertical distribution of dust aerosols  
165 (Amiridis et al., 2013) or specifying the vertical structure of dust events (Amiridis et al., 2009).  
166 Nevertheless, they only dealt with a single dust event (18-23 May 2008, Amiridis et al., 2009) and thus  
167 cannot satisfy the need to know the general vertical structure of Mediterranean dust episodes.

168 The main target of the present study is to describe the Mediterranean desert dust outbreaks' vertical  
169 structure. For this purpose, satellite retrievals derived by the MODIS-Terra/Aqua, EP-TOMS, OMI-  
170 Aura and CALIOP-CALIPSO databases (Section 2) are used in a synergistic way. The dust outbreaks  
171 are identified with an objective and dynamic algorithm, which uses appropriate aerosol optical  
172 properties representative of suspended particles' load, size and nature (Section 3). First, the outputs of  
173 the satellite algorithm are compared versus surface measurements provided by AERONET or  $PM_{10}$   
174 stations, located within the study region (Section 4.1). Additionally, useful information about various  
175 optical and physical properties under intense dust episodes conditions is also derived from the  
176 aforementioned analysis. Then, the primary characteristics of the intense Mediterranean desert dust  
177 (DD) episodes, namely their frequency and intensity, are described in Section 4.2. Just in order to  
178 assess the consistency of the algorithm' concept, an alternative methodology for the determination of  
179 DD episodes is also applied and the obtained results are inter-compared with the basic methodology.  
180 For the identified DD episodes, collocated CALIOP-CALIPSO vertical feature mask and total  
181 backscatter coefficient at 532 nm retrievals are used in order to describe the annual and seasonal  
182 variability of dust outbreaks' vertical extension over the Mediterranean (Section 4.3). Moreover, in  
183 Section 4.4, a thorough analysis of specific Mediterranean DD episodes is made, in order to examine  
184 how the vertical distribution of desert dust outbreaks can affect the agreement between MODIS  $AOD$   
185 and  $PM_{10}$  data. Finally, the summary and conclusions are drawn in Section 5.

186

## 187 2. Satellite and surface-based data

188 The different types of satellite retrievals that have been used as inputs to the objective and dynamic  
189 satellite algorithm are described below, namely the MODIS (Section 2.1.1), EP-TOMS and OMI-Aura  
190 (Section 2.1.2) databases. Also, CALIOP-CALIPSO vertically resolved satellite data, coincident with  
191 the identified desert dust outbreaks by the satellite algorithm, are described in Section 2.1.3. Finally,

192 surface-based sun-photometric AERONET retrievals and  $PM_{10}$  concentrations, both used for the  
193 comparison against the satellite algorithm's outputs, are described in Sections 2.2.1 and 2.2.2,  
194 respectively.

195

## 196 2.1 Satellite data

### 197 2.1.1 MODIS

198

199 MODerate resolution Imaging Spectroradiometer (MODIS) onboard the Terra and Aqua satellites –  
200 with daytime local equator crossing time at 10:30 and 13:30 UTC, respectively, and 2330 km viewing  
201 swath – acquires measurements at 36 spectral bands between 0.415 and 14.235  $\mu\text{m}$  with varying spatial  
202 resolution of 250, 500 and 1000 m. Observations from Terra and Aqua are made continuously since  
203 February 2000 and July 2002, respectively, and are available from the LAADS website  
204 (<ftp://ladsweb.nascom.nasa.gov/>). Aerosol optical properties are retrieved through the Dark Target  
205 (DT) algorithm (see e.g. Kaufman et al., 1997, 2001; Tanré et al., 1997; Levy et al., 2003; Remer et al.,  
206 2005) where different assumptions are considered depending on the underlying surface type (land or  
207 ocean). Several evaluation studies (e.g. Remer et al., 2008; Papadimas et al., 2009; Levy et al., 2010;  
208 Nabat et al., 2013) have shown that aerosol optical depth ( $AOD$ ) can be retrieved satisfactorily by  
209 MODIS, nevertheless its performance is better over sea (uncertainty equal to  $\pm 0.03 \pm 0.05 \times AOD$ ;  
210 Remer et al., 2002) than over land ( $\pm 0.05 \pm 0.15 \times AOD$ ; Levy et al., 2010).

211 The following daily MODIS-Terra and MODIS-Aqua Collection 051 (C051) level 3 satellite data  
212 (MOD08\_D3 and MYD08\_D3 files) provided at  $1^\circ \times 1^\circ$  latitude-longitude spatial resolution are used:  
213 (i)  $AOD_{550nm}$ , (ii) Ångström exponent over land ( $\alpha_{470-660nm}$ ), (iii) Ångström exponent over ocean  
214 ( $\alpha_{550-865nm}$ ), (iv) fine-mode fraction ( $FF$ ) of  $AOD$  over land and ocean and (v) Effective radius over  
215 ocean ( $r_{eff}$ ). It must be mentioned that the size parameters ( $\alpha$ ,  $FF$ ) over land are less reliable compared  
216 to the corresponding ones over sea, since they are highly sensitive to spectral dependent factors such as  
217 errors in the surface model or sensor calibration changes. Over sea, the accuracy of size parameters is  
218 strongly dependent on wind conditions.

219 Similar data have been used by Gkikas et al. (2013). However, in the present study we have  
220 improved data quality by using the quality assurance-weighted (QA) level 3 data ([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)) derived from the level 2 retrievals (10 km x 10

222 km spatial resolution). Each level 2 retrieval, is flagged with a bit value (from 0 to 3) corresponding to  
223 confidence levels (No confidence: 0, Marginal: 1, Good: 2 and Very Good: 3). Based on this, the level  
224 3 QA-weighted spatial means are obtained by the corresponding level 2 retrievals considering as  
225 weight their confidence level (bit value). In addition, the day cloud fraction as well as the number of  
226 level 2 counts, which are both relevant to the performance of the satellite algorithm, are also used in  
227 this study. The time series of daily MODIS aerosol data cover the 13-yr period March 2000-February  
228 2013 (Terra) and the 10-yr period January 2003-December 2012 (Aqua).

229

### 230 2.1.2 EP/TOMS and OMI-Aura

231 The selected retrievals from MODIS provide information about particles' load (*AOD*) and size ( $\alpha$ ,  
232  $FF$ ,  $r_{eff}$ ), which are both necessary to identify dust episodes. However, since dust is not the only coarse  
233 aerosol, for example sea-salt can be so as well, another optical property indicative of particle  
234 absorption efficiency is also required by the algorithm. To address this issue, the Absorption Aerosol  
235 Index (*AI*) daily data were also used, derived from measurements taken by the Total Ozone Mapping  
236 Spectrometer (TOMS) instrument onboard the NASA's Earth-Probe satellite (2000-2004) and the  
237 Ozone Monitoring Instrument (OMI) onboard the NASA's Aura satellite (2005-2013). *AI* is the  
238 primary TOMS aerosol product (Herman et al., 1997) based on a spectral contrast method in a UV  
239 region (331-360 nm) where ozone absorption is very small and can be used for the distinction between  
240 scattering (e.g. sea-salt) and absorbing (e.g. desert dust, smoke) aerosols. The retrieval algorithm (fully  
241 described by Torres et al., 1998; 2002; 2005) takes advantage of the low surface albedo in the UV  
242 spectrum range, even in arid and semi-arid areas, making thus possible the estimation of the *AOD* over  
243 highly reflecting desert surfaces, where the major dust sources are located. Since the late 70's, the  
244 TOMS sensor onboard Nimbus-7 (1978 – 1993) and Earth Probe (1996 – 2005) has been providing  
245 global aerosol measurements. With the deployment of the EOS-Aura OMI (Ozone Monitoring  
246 Instrument) in mid-2004 (Torres et al., 2007) the near UV aerosol record continues to be extended into  
247 the foreseeable future. OMI is a hyperspectral sensor, covering the 270-500 nm range, launched  
248 onboard the EOS-Aura satellite on July 15, 2004 (1:38 pm equator crossing time, ascending mode)  
249 providing almost daily global coverage thanks to its wide viewing swath (2600 km with 13 km x 24 km  
250 nadir resolution). Apart from *AI* measurements, OMI aerosol products include also the total and  
251 absorption *AOD* and the single scattering albedo at 388 and 500 nm (Torres et al., 2007). Both EP-  
252 TOMS and OMI-Aura retrievals are available via the Mirador ftp server (<http://mirador.gsfc.nasa.gov/>)



253 of the Goddard Earth Sciences Data and Information Services Center (GES DISC). OMI-Aura data, as  
254 MODIS, are provided at  $1^\circ \times 1^\circ$  spatial resolution while the EP-TOMS retrievals have been regridded  
255 from their raw spatial resolution ( $1^\circ \times 1.25^\circ$ ) in order to match the other two datasets (OMI, MODIS).

256

### 257 2.1.3 CALIOP-CALIPSO

258

259 The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard the NASA's satellite  
260 CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations), launched in April  
261 2006, provides vertical resolved aerosol and cloud observations (Winker et al., 2009) since June 2006.  
262 CALIPSO is flying in the A-Train constellation (Stephens et al., 2002; <http://atrain.nasa.gov/>) in a sun-  
263 synchronous polar orbit at 705 km over the surface, with a 16-day repeat cycle, crossing the equatorial  
264 plane at about 13:30 local solar time (Winker et al., 2009). CALIOP is an active sensor measuring the  
265 backscatter signal at 532 nm and 1064 nm as well as the polarization at 532 nm (Winker et al., 2009).  
266 These level 1 retrievals are further processed (calibration and range corrections) passing to level 2 in  
267 order to retrieve the backscatter and extinction coefficients, at 532 nm and 1064 nm, for aerosol and  
268 cloud layers. The identification of cloud and aerosol layers within the atmosphere (Vaughan et al.,  
269 2009) is made through the cloud aerosol discrimination (CAD) algorithm (Liu et al., 2009), which is  
270 based on the probability distribution functions (PDFs) of altitude-and-latitude-dependent parameters  
271 (integrated color ratio, layer-integrated volume depolarization ratio, mean attenuated backscatter  
272 coefficient). CAD scores vary mainly from -100 to 100 indicating the presence of aerosols and clouds  
273 when negative and positive, respectively, while bins of confidence levels, both for aerosols and clouds,  
274 are defined based on their absolute values  
275 ([https://eosweb.larc.nasa.gov/sites/default/files/project/calipso/quality\\_summaries/CALIOP\\_L2VFMPr](https://eosweb.larc.nasa.gov/sites/default/files/project/calipso/quality_summaries/CALIOP_L2VFMProducts_3.01.pdf)  
276 [oducts\\_3.01.pdf](https://eosweb.larc.nasa.gov/sites/default/files/project/calipso/quality_summaries/CALIOP_L2VFMProducts_3.01.pdf)). More specifically, the performance of the classification scheme in the retrieval  
277 algorithm, either for aerosols or clouds, is more reliable for increasing CAD scores in absolute terms.  
278 Aerosols are categorized in 6 primary types, namely: (i) clean marine, (ii) dust, (iii) polluted  
279 continental, (iv) clean continental, (v) polluted dust and (vi) smoke (Omar et al., 2009).

280 In the present analysis, we use the Version 3 (3.01 and 3.02) of the Level 2 Vertical Feature Mask  
281 (VFM) and Aerosol Profile Products (APro) files, available from June 2006 to February 2013, both  
282 derived from the NASA's Earth Observing System Data and Information System  
283 (<http://reverb.echo.nasa.gov/>). The aerosol profile products are generated at a uniform horizontal  
284 resolution of 5 km ([http://www-calipso.larc.nasa.gov/products/CALIPSO\\_DPC\\_Rev3x6.pdf](http://www-calipso.larc.nasa.gov/products/CALIPSO_DPC_Rev3x6.pdf)), while the

vertical resolution varies from 60 to 180 m depending on the altitude range and the parameter. The scientific data sets which have been analyzed are the following: (i) aerosol subtype, (ii) *CAD* score and (iii) Total Backscatter Coefficient at 532 nm ( $\beta_{532nm}$ ), reported at several tropospheric and stratospheric levels above mean sea level (Hunt et al., 2009).

## 2.2 Surface-based data

### 2.2.1 AERONET

The AErosol RObotic NETwork (AERONET, Holben et al., 1998) is a worldwide network of installed CIMEL sun-sky radiometers obtaining sun-photometric observations in more than 1000 locations of the planet (<http://aeronet.gsfc.nasa.gov>). The solar irradiances received by the photometer are inverted into 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 ( $\alpha_{440-870nm}$ ), (iv) total effective radius ( $r_{eff}$ ), and (v) single scattering albedo (*SSA*) and asymmetry parameter ( $g_{aer}$ ) both retrieved at 440 nm, 675 nm, 870 nm 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 better ( $< \pm 0.01$ ) for wavelengths longer than 440 nm and lower ( $< \pm 0.02$ ) for the 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).

### 316 2.2.2 $PM_{10}$

317 Daily total and dust surface  $PM_{10}$  concentrations, over the period 2001-2011 from 22 regional  
318 background and suburban background sites were used in this study. The monitoring sites are distributed  
319 as follows: 10 in Spain; 2 in southern France; 5 in Italy; 3 in Greece; 1 in southern Bulgaria and 1 in  
320 Cyprus.  $PM_{10}$  concentrations were obtained in most cases from gravimetric determinations on filters,  
321 whereas in few cases they were determined by real time instruments (Querol et al., 2009b; Pey et al.,  
322 2013) but corrected against gravimetric measurements carried out in annual field campaigns. The  
323 disaggregation of the dust component to the total amount is made based on a statistical approach which  
324 has been applied in several past studies (e.g. Rodríguez et al., 2001; Escudero et al., 2007; Querol et al.,  
325 2009b; Pey et al., 2013). A full description of the methodology which is followed for the calculation of  
326 dust particles' contribution to the total  $PM_{10}$  is presented in Escudero et al. (2007). Briefly, the net dust  
327  $PM_{10}$  amount is calculated through the subtraction of the regional background  $PM_{10}$ , which is obtained  
328 by applying a monthly moving 30<sup>th</sup> percentile to the  $PM_{10}$  timeseries excluding days of dust transport,  
329 from the corresponding values of the total  $PM_{10}$  concentrations. Most of the derived data were obtained  
330 from the AirBase (<http://acm.eionet.europa.eu/databases/airbase/>) database, while for the stations  
331 Finokalia (Crete) and Montseny (NE Spain) the relevant measurements have been acquired from the  
332 EUSAAR (<http://www.eusaar.net/>) database.

333

### 334 3. Identification of desert dust episodes

335

336 Following the methodology proposed by Gkikas et al. (2013), desert dust (DD) episodes are  
337 identified based on an objective and dynamic algorithm which consists a branch of a unified algorithm  
338 (Gkikas et al., 2016) able to identify and characterize not only DD episodes, but also four other types of  
339 aerosol episodes, namely biomass-urban (*BU*), dust/sea-salt (*DSS*), mixed (*MX*) and undetermined  
340 (*UN*). The algorithm (see Figure 2 in Gkikas et al., 2013) operates in three steps and is applied in each  
341 individual 1° x 1° geographical cell within the geographical limits of the study domain (29° N - 47° N  
342 and 11° W - 39° E). First, the mean (*Mean*) and the associated standard deviation (*Std*) from the  
343 available  $AOD_{550nm}$  retrievals are calculated for the whole study period. These primary statistics are  
344 used for the definition of two threshold levels, which are equal to  $Mean+2*Std$  and  $Mean+4*Std$ . The  
345 geographical distributions of the computed statistics (*Mean* and *Std*) as well as the corresponding  
346 spatial patterns of both threshold levels are displayed in Figures S1-a (MODIS-Terra, Mar. 2000 – Feb.  
347 2013) and S1-b (MODIS-Aqua, 2003 – 2012) in the supplementary material. At the next step, the

algorithm analyzes the daily  $AOD_{550nm}$  timeseries and classifies an episode as a strong one when  $AOD$  is between the two defined thresholds ( $Mean+2*Std \leq AOD_{550nm} < Mean+4*Std$ ) and as an extreme one when  $AOD$  is higher/equal than  $Mean+4*Std$ . The same approach was undertaken by Gkikas et al. (2009) who classified the Mediterranean aerosol episodes over the period 2000-2007 according to their strength and described their frequency and intensity. It must be clarified that according to our methodology in areas frequently affected by dust episodes, both mean and standard deviation values are expected to be high resulting to high thresholds which means that cases with moderate-to-high  $AODs$ , also possibly relevant to radiative and health effects, are masked out from the dataset. In order to investigate the possible impact of this, “unbiased” mean, standard deviation and thresholds of  $AOD$  are also computed based on another methodology and the results are discussed comparatively to those of the primary methodology in a separate paragraph. Moreover, it must be mentioned that the satellite algorithm identifies only intense desert dust episodes since their  $AOD$  must be higher than  $Mean+2*Std$  which is considered as a high threshold level.

It should be noted that the representativeness of the calculated mean levels is possibly affected by the availability of the  $AOD$  retrievals and particularly by the way these data are distributed both at temporal and spatial scales. Thus, a possible underrepresentation of winter  $AOD$  data in the long-term dataset, which is often the case in satellite retrievals of  $AOD$ , may result in a higher mean  $AOD$  than what would be in case of complete and balanced seasonal availability. Moreover, the spatiotemporal availability of  $AOD$  is determined by the different satellite retrieval algorithm assumptions depending on the underlying surface type (land or sea) and clouds (i.e. satellite retrievals are possible only under clear skies conditions). In order to investigate the possible effect of temporal availability of daily  $AOD$  data, we have calculated the percentage availability of  $AOD$  retrievals on a monthly, seasonal and year by year basis, over the period 2000-2013 (results not shown here). Seasonal differences of  $AOD$  availability are mainly encountered in the northernmost parts of the study region, attributed to the enhanced cloud coverage, with lower values (20 to 40 %) from December to February against 50-85% for the rest of the year. Differences in  $AOD$  availability are also found between land and sea surfaces which are more pronounced in winter and summer and less remarkable during the transition seasons. More specifically, across the Mediterranean Sea, in winter, the availability percentages range from 70 to 90 % while in summer the corresponding values are decreased, due to Sun glint, down to 60 % and 80 %, respectively. Over land, for both seasons, the spatial patterns of  $AOD$  availability are reversed. In order to investigate furthermore how the spatiotemporal  $AOD$  variability and unbalanced seasonal distribution of MODIS  $AOD$  data can affect the calculated mean  $AOD$  levels (calculated by daily

380 retrievals), we have repeated the calculations by utilizing monthly retrievals (calculated from the daily  
381 ones) thus removing the possible effects of an unequal temporal distribution of the number of  
382 observations on the mean *AOD*. According to our results, only small differences are found, generally  
383 hardly exceeding 0.1 in absolute and 5% in relative percentage terms, with the mean *AODs* over land  
384 being higher by up to 10 % when they are computed from daily than monthly data, while the opposite is  
385 found over sea. This finding reveals that the unequal temporal distribution of *AOD* retrievals does not  
386 have critical impact on the computed mean *AODs* and the resulting algorithm outputs presented in this  
387 study.

388 In a further step of the methodology, the strong and extreme DD episodes are identified separately  
389 over land and sea surfaces of the study region. This is achieved through the usage of specific aerosol  
390 optical properties, namely the Ångström exponent, effective radius, fine fraction and aerosol index,  
391 which provide information about particles' size and nature. For each optical property, appropriate  
392 upper or lower thresholds have been set up which must be valid concurrently in order to certify the  
393 presence of dust particles in the atmosphere. Note that there are not any unanimously defined  
394 acknowledged thresholds in literature. Therefore, these cut-off levels have been selected here according  
395 to the literature findings, availability of raw data and several own sensitivity tests (more details are  
396 provided in Gkikas et al., 2013) which have been applied individually to the MODIS size parameters  
397 (i.e.,  $\alpha$ , *FF* and  $r_{eff}$ ). Such analysis is essential when multi-parameter datasets are utilized and their  
398 variations can possibly modify the satellite algorithm's outputs. To this aim, we have applied the  
399 satellite algorithm modifying by 0.1 the  $\alpha$ , *FF* and  $r_{eff}$  values within the ranges 0.6 – 0.8, 0.1 – 0.4 and  
400 0.4 – 0.8, respectively. Our results indicate that the geographical patterns remain similar and the total  
401 number of DD episodes is only slightly modified (less than 4 %) for the  $\alpha$  and  $r_{eff}$  retrievals, whereas it  
402 changes more for the *FF* retrievals (by up to 25% over sea for strong episodes). Here, the validity of  
403 these thresholds is further evaluated against AERONET measurements and the corresponding results  
404 are discussed in Section 4.1.1.4.

405 In order to address the issue of possible overestimation of the defined threshold levels, particularly  
406 in the most dust-affected areas as it has been mentioned above, we have also applied the satellite  
407 algorithm using an alternative methodology (METHOD-B) in which dust-affected grid cells were  
408 excluded. In this case, from the raw *AOD* retrievals we have masked out the “pure” desert dust grid  
409 cells, which were identified based on the concurrent fulfillment of the defined criteria for dust  
410 occurrence in the algorithm (for Ångström exponent, fine fraction, aerosol index and effective radius).  
411 Then, from the remaining data (non-dust *AOD* retrievals), the mean, the associated standard deviation

412 as well as the defined thresholds of *AOD* are computed for the whole study period, for each pixel, as  
413 also done in the primary methodology. Finally, also similarly to the way done in the primary  
414 methodology, the DD episodes were classified into strong and extreme ones. The obtained results, i.e.  
415 frequency of occurrence and intensity of DD episodes, based on the primary methodology and  
416 METHOD-B are discussed in Section 4.2.

417 As explained, a similar methodology and data were used in the study by Gkikas et al. (2013).  
418 Nevertheless, the present one is a significant extension mainly for five reasons: (i) DD episodes are  
419 identified here over an extended period of study and for both MODIS platforms, i.e. Mar. 2000 – Feb.  
420 2013 for MODIS-Terra and 2003 – 2012 for MODIS-Aqua, (ii) a second methodology (METHOD-B)  
421 for the identification of DD episodes is tested, (iii) the quality of the input data is improved by using  
422 QA-weighted level-3 data produced by weighting level-2 data based on their confidence flag instead of  
423 regular ones, (iv) emphasis is given to the vertical structure of the intense DD episodes and (v) the role  
424 of the detailed dust outbreaks' vertical structure for the level of agreement between columnar MODIS  
425 *AOD* and ground  $PM_{10}$  concentrations is investigated. In addition, in the present analysis, the satellite  
426 algorithm is also tested using only *AODs* associated with cloud fractions (*CF*) lower/equal than 0.8, in  
427 order to investigate possible modifications of our results due to the cloud contamination effects on  
428 MODIS *AODs*. The critical value of 0.8 for *CF* has been defined according to Zhang et al. (2005) and  
429 Remer et al. (2008), who stated that under extended cloud coverage conditions *AOD* levels can be  
430 increased substantially.

431

## 432 **4. Results**

433 Before dealing with the horizontal patterns (sub-section 4.2) and the vertical structure of dust  
434 outbreaks (sub-sections 4.3 and 4.4), it is very important to compare the algorithm's outputs against  
435 AERONET and  $PM_{10}$  observations (sub-section 4.1) in order to ensure an accurate three-dimensional  
436 view of the intense Mediterranean DD episodes. It must be clarified that the comparison of the satellite  
437 algorithm's outputs versus AERONET/ $PM_{10}$  is made only for its default version and not for the  
438 METHOD-B, since between the two methodologies are not found remarkable differences, as it will be  
439 presented in Section 4.2. For the same reason, the synergistic implementation of the CALIOP-  
440 CALIPSO lidar profiles is done only when the DD episodes are identified based on the primary  
441 methodology. The present section has been organized accordingly and the results are given below.

442

#### 4.1 Comparison of the satellite algorithm's outputs against AERONET and $PM_{10}$ measurements

The ability of the satellite algorithm to identify satisfactorily DD episodes, is tested against ground measurements from 109 AERONET (Fig. 1, orange squares) and 22  $PM_{10}$  (Fig. 1, green triangles) stations located in the broader Mediterranean area. This is an extended and thorough comparison which exceeds largely a similar one done for the outputs of the previous version of satellite algorithm (2000-2007; Gkikas et al., 2013), but only relying on 9 AERONET stations and using *AOD* and volume size distribution data. Here, the comparison is repeated for the improved algorithm, being extended over a longer time period, for a much larger number of AERONET stations, and an analysis of more optical properties, namely the Ångström exponent, effective radius, single scattering albedo and asymmetry parameter is made. The comparison is performed for both study periods and satellite platforms (Mar. 2000 – Feb. 2013 for Terra and 2003 – 2012 for Aqua) 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 Mar. 2000 – Feb. 2013 are given here.

In 46 out of 109 AERONET stations, depicted with yellow triangles in Figure 1, we have 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) 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 Mar. 2000 – Feb. 2013, aiming at highlighting the effect of episodes on these optical properties. Additionally, in 7 AERONET stations (cyan circles in Figure 1) the intense DD episodes have been identified from ground (AERONET) and the corresponding results are compared with the satellite algorithm outputs (Section 4.1.1.4). Finally, the performance of the algorithm is also tested against surface  $PM_{10}$  measurements from 22 stations (Section 4.1.2).

#### 473 4.1.1 AERONET

##### 474 4.1.1.1 Aerosol optical depth

475 During the period Mar. 2000 – Feb. 2013, 346 pixel-level intense DD episodes have been identified  
476 by the satellite-based algorithm, in which coincident MODIS-Terra and AERONET retrievals are  
477 available. The selected dataset corresponds to 1.06 % of the overall (strong and extreme) DD episodes  
478 (32635) which have been identified during the study period. It should be noted that AERONET  
479  $AOD_{550nm}$  values have been calculated from available AERONET  $AOD_{870nm}$  and Ångström exponent  
480 data ( $\alpha_{440-870nm}$ ) by applying the Ångström equation (Ångström, 1929) to match the MODIS  $AOD_{550nm}$ .  
481 For these intense DD episodes, the comparison between the satellite and ground aerosol optical depths  
482 at 550 nm is given in Figure 2. Two similar scatterplots with matched MODIS-AERONET data pairs  
483 are given. The first one (Fig. 2 i-a) is resolved by the number of level 2 ( $L2$ ) measurements of 10 km x  
484 10 km spatial resolution from which the compared  $1^\circ \times 1^\circ$  level 3 ( $L3$ )  $AODs$  in the figure are derived.  
485 The second scatterplot (Fig. 2 i-b) is resolved by the spatial standard deviation inside the  $1^\circ \times 1^\circ$   
486 geographical cell (level 3  $AODs$ ). Both scatterplots address the issue of level 3  $AOD$  sub-grid spatial  
487 variability, which is essential when attempting comparisons against local surface-based  $AOD$  data like  
488 the AERONET.

489 The overall correlation coefficient ( $R$ ) between MODIS and AERONET  $AODs$  is equal to 0.505,  
490 with the satellite  $AODs$  being overestimated (bias=0.143). The scatterplots show the existence of  
491 outliers associated with small number of level 2 retrievals ( $< 20$ , blue color Fig. 2 i-a) and/or high  
492 standard deviations ( $> 0.5$ , yellowish-reddish points, Fig. 2 i-b) inside the  $L3$  grid cell. This finding  
493 underlines the role of homogeneity and representativeness of  $L3$  retrievals for the comparison of  
494 MODIS  $AODs$  against AERONET. This role is better visualized in Fig. 2 ii-a, where are presented the  
495 computed  $R$  values between MODIS level-3 and AERONET  $AODs$  depending on the number of  $L2$   
496 retrievals from which the  $L3$  products were derived. In general, it is known that the  $L2$  pixel counts  
497 range from 0 to 121 while in polar regions (typically around  $82^\circ$  latitude) the maximum count numbers  
498 can be even higher due to overlapping orbits and near nadir views intersect (Hubanks et al., 2008). It is  
499 clear from our results that the correlation coefficients are gradually and essentially improved, from 0.49  
500 to 0.75, with increasing representativeness of MODIS  $AODs$ , i.e. increasing counts of  $L2$  retrievals  
501 attributed. A similar improvement has been reported by Amiridis et al. (2013) who found a better  
502 agreement between MODIS/AERONET and CALIOP aerosol optical depths applying similar criteria.  
503 The agreement between MODIS and AERONET also improves when the former  $AOD$  products are



504 more spatially homogeneous, i.e. when they are characterized by smaller *AOD* standard deviations at  
505 the grid-level (from  $< 0.25$  down to  $< 0.05$ , Fig. 2 ii-b). However, our results also indicate that apart  
506 from increasing correlation coefficients (up to 0.7-0.8) with increasing level-2 counts and decreasing  
507 standard deviations, the number of intense DD episodes is decreased dramatically (about 40-50 for  
508 more than 50 counts and standard deviation smaller than 0.05).

509 In order to assess the performance of the satellite algorithm when operated with non-weighted  
510 (Gkikas et al., 2013) and weighted QA (present analysis) MODIS-Terra retrievals we have compared  
511 its outputs (DD episodes' *AODs*) of both versions versus the corresponding AERONET *AODs* for the  
512 period Mar. 2000 – Feb. 2007 (Gkikas et al., 2013). Based on our results, the computed correlation  
513 coefficients are equal to 0.53 (135 DD episodes) and 0.59 (177 DD episodes) for the old and new  
514 version of the satellite algorithm, respectively, revealing thus a better performance when QA-weighted  
515 level 3 retrievals are utilized as inputs to the satellite algorithm.

516 Finally, the spectral variation of the AERONET *AODs* at 7 wavelengths, from 340 to 1020 nm, in  
517 climatological and dust episodes conditions has been investigated (results given in Figure S2,  
518 supplementary material). The *AOD* boxplots produced for all the available daily AERONET  
519 measurements (orange) and for the corresponding retrievals during strong (cyan), extreme (red) and all  
520 DD (green) episodes identified by the satellite algorithm show that the spectral variation of aerosol  
521 optical depth decreases in cases of dust episodes, with respect to the “climatological” conditions. This  
522 is mainly attributed to the further increasing *AOD* levels at wavelengths longer than 500 nm (by about  
523 6 times) than in (or near) the visible.

524

#### 525 4.1.1.2 Aerosol volume size distribution

526 In Figure 3, are presented the mean aerosol volume size distributions (*AVSDs*) calculated from all  
527 available AERONET data (orange curve) as well as under strong (cyan curve), extreme (red curve) and  
528 all (green curve) DD episodes conditions. The results are given for Mar. 2000 – Feb. 2013 using  
529 MODIS-Terra (346 intense DD episodes) retrievals as inputs to the satellite algorithm. In the  
530 climatological curve, two modes are distinct centered at  $0.15\ \mu\text{m}$  for the fine mode and  $2.24\ \mu\text{m}$  for the  
531 coarse mode. There is an about equal contribution of both modes, indicating the coexistence of fine  
532 (e.g. urban aerosols) and coarse (e.g. dust aerosols) particles over the broader Mediterranean area. This  
533 result is in agreement with previous studies for the Mediterranean (e.g. Fotiadi et al., 2006; Mallet et  
534 al., 2013). However, under dust episodes conditions, although the *AVSD* still has two modes, there is a

dramatic increase of the coarse mode, which strongly dominates. More specifically, the peak of the coarse mode (radius between 1.7 and 2.24  $\mu\text{m}$ ) is increased by factors of about 10, 15 and 11 for the strong, extreme and all DD episodes. The differences between the strong and extreme AVSDs are statistically significant (confidence level at 95 %) for almost all size bins (18 out of 22) except bin 1 (0.050  $\mu\text{m}$ ), 2 (0.065  $\mu\text{m}$ ), 6 (0.194  $\mu\text{m}$ ) and 7 (0.255  $\mu\text{m}$ ). Moreover, it should be noted that the increment factors are slightly decreased when the algorithm operates only with AOD retrievals associated with cloud fractions less than 0.8 which is reasonable since possible “overestimated” retrievals are masked out from the analysis. Similar modifications in the shape of AVSD during dust outbreaks have been pointed out by several studies in the past, either for the Mediterranean region (e.g. Kubilay et al., 2003; Lyamani et al., 2005; Córdoba-Jabonero et al., 2011) or for other dust affected areas of the planet (e.g. Alam et al., 2014; Cao et al., 2014).

#### 4.1.1.3 Size optical properties, single scattering albedo and asymmetry parameter

The accuracy of the DD episodes identification method was further assessed by also using other AERONET aerosol optical properties than AOD, namely the Ångström exponent ( $\alpha$ ) and the effective radius ( $r_{\text{eff}}$ ), able to provide information about particles’ size. For both aerosol optical properties, the boxplots for all the available AERONET retrievals as well as for the corresponding data during strong, extreme and all DD episodes, have been produced and depicted in Figure S3 (supplementary material).

Based on our results, the appropriateness of the applied methodology is confirmed by the drastic reduction of  $\alpha$  and increase of  $r_{\text{eff}}$  values when dust outbreaks occur. When all available AERONET retrievals are considered (*clim*), the majority (> 75%) of  $\alpha$  values is higher than 1.04 indicating the strong presence of fine particles in the study domain (Figure S3-i). On the contrary, during intense dust episodes the majority of the corresponding values for all and strong DD episodes are lower than 0.54 while for the extreme ones are lower than 0.36. Such low Ångström exponent values, attributed to transported mineral particles from the northern African deserts (Pace et al., 2006), have been reported also in previous studies (e.g. Tafuro et al. 2006; Basart et al., 2009). The existence of coarse aerosols is also confirmed by the increase of  $r_{\text{eff}}$  values under intense DD conditions compared to the climatological levels (Figure S3-ii). For all DD episodes, the 75% of  $r_{\text{eff}}$  values is higher than 0.55  $\mu\text{m}$  reaching up to 1.4  $\mu\text{m}$ , while the mean and the median values are equal to about 0.73, compared to about 0.37 for the climatological conditions. These values are even higher when extreme DD episodes are concerned.

Moreover, the spectral variations of the averaged AERONET single scattering albedo ( $SSA$ ) and the asymmetry parameter ( $g_{aer}$ ) are also studied. During intense dust outbreaks the shape and magnitude of spectral  $SSA$  (Figure S4-i) and  $g_{aer}$  (Figure S4-ii) are modified compared to the climatological conditions. The spectral curves of both parameters become less and more flattened during dust episodes for  $SSA$  and  $g_{aer}$ , respectively. For  $SSA$ , the steepening results from decreasing values in the visible and increasing values in the near-infrared (by up to 0.04, reaching 0.97 at 1020 nm). The flattening for  $g_{aer}$  arises from smaller and larger increments in visible and near-infrared wavelengths, by up to 0.04 and 0.09, respectively. The differences between strong and extreme DD episodes  $SSA$  spectral curves are statistically significant at 95 % confidence level only at 870 and 1020 nm. On the contrary, the corresponding differences for the  $g_{aer}$  are statistically significant in all wavelengths. Our results are in agreement with those presented for  $SSA$  by Mallet et al. (2013) in the Mediterranean and for  $g_{aer}$  by Alados-Arboledas et al. (2008) during a dust episode over the southeastern parts of Spain.

578

#### 4.1.1.4 Intercomparison of surface-based and satellite algorithms used for the identification of the desert dust episodes

Despite their great usefulness, satellite aerosol retrievals still suffer from uncertainties, and generally are considered as inferior to surface-based similar products, which are taken as the reference. In order to examine this degree of uncertainty and to verify the successful performance of the algorithm, we also tested using it along with AERONET retrievals. This has been made for 7 Mediterranean AERONET stations, depicted with cyan circles in Figure 1, during the periods for which ground retrievals are available (Table 1). The selection of the AERONET stations was based on: (i) data availability (see last column of Table 1), (ii) their location (i.e. near to the Northern African and Middle East deserts) and (iii) the inclusion of sites where the aerosols' regime is complex (e.g. El Arenosillo, FORTH Crete). The intense DD episodes were identified following the methodology described in section 3, but using only  $AOD$  at 870 nm,  $\alpha_{440-870nm}$  (lower/equal than 0.7) and  $r_{eff}$  (higher than 0.6) as criteria, based upon their availability from AERONET. Subsequently, the algorithm was also operated again using satellite (MODIS-Terra, OMI-Aura, EP-TOMS) input data for the periods with available retrievals in each of the 7 AERONET stations.

In Figure 4, we present 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 Figs. 4 i-a, 4 ii-a, 4 iii-a represent the associated MODIS-Terra Ångström

597 exponent, effective radius and day cloud fraction retrievals, respectively. In Figs. 4 i-b and 4 ii-b colors  
598 represent the AERONET Ångström exponent and effective radius, respectively, while in Figure 4 iii-b  
599 they represent the day cloud fraction observations derived by MODIS-Terra. Through this approach it  
600 is feasible to assess furthermore the performance of the satellite algorithm, specify its drawbacks and  
601 check the validity of the defined thresholds (green boxes in Figure 2 in Gkikas et al., (2013)).

602 It is apparent that the agreement between MODIS-Terra and AERONET *AODs* is better when DD  
603 episodes are identified from the ground, as shown by the increased correlation coefficients (from 0.521  
604 to 0.704), increased slopes (from 0.6 to 0.9-1.0) and decreased biases (from 0.16 to -0.03). In  
605 particular, when DD episodes are identified from space, the MODIS-Terra *AOD* retrievals are  
606 overestimated (bias=0.163) with regards to AERONET, particularly at low *AOD* values (< 0.5). In both  
607 algorithms, the highest overestimations are associated with cloud fractions higher than 0.7 due to the  
608 possible contamination of the satellite *AODs* by clouds (Figure 4 iii-a, iii-b). Given that DD episodes'  
609 identification based on AERONET retrievals is more efficient, we have used these results in order to  
610 check the validity of the defined thresholds for  $\alpha$ , *AI*, *FF* and  $r_{eff}$  used in the satellite algorithm. For  
611 each aerosol optical property, it has been calculated the percentage of intense DD episodes for which  
612 the corresponding satellite observations are below or above the defined thresholds, depending on the  
613 parameter. The results given in Table 2 are satisfactory, since the percentages range from 87 to 99%,  
614 and confirm the validity of the defined thresholds.

615 The scatterplots in Figs. 4 i-b and ii-b also reveal some weaknesses of the satellite-based algorithm.  
616 More specifically, it is found that for few DD episodes identified by the satellite algorithm the  
617 corresponding AERONET Ångström exponent and effective radius values are higher than 1 and  
618 smaller than 0.4, respectively. These values indicate a predominance of fine particles instead of coarse  
619 ones as it would be expected for desert dust aerosols. In order to quantify the number of misclassified  
620 pixel level intense DD episodes by the satellite algorithm, we have computed the percentage of cases  
621 for which the AERONET  $\alpha$  values are higher than 1 (15%) and  $r_{eff}$  values are lower than 0.4 (17.7%).  
622 Also, we have repeated these calculations for all DD episodes and the corresponding percentages were  
623 found to be equal to 11.8% and 14.5%, respectively. These misclassifications of the satellite algorithm  
624 occur in AERONET stations (e.g. Thessaloniki, Rome, Avignon) with a strong presence of  
625 anthropogenic aerosols (Kazadzis et al., 2007; Gobbi et al., 2007; Querol et al., 2009a; Yoon et al.,  
626 2012). Some misclassifications also occur in AERONET stations (e.g. Evora, El Arenosillo, FORTH  
627 CRETE) with mixed (natural plus anthropogenic) aerosol loads (Fotiadi et al., 2006; Toledano et al.,  
628 2007b; Hatzianastassiou et al., 2009; Pereira et al., 2011). Over these areas, there are converging air

629 masses carrying particles of different origin, as shown by performed back-trajectories analyses (results  
630 are not shown here) using the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory)  
631 model (Draxler and Rolph, 2015). Nevertheless, it must be mentioned that DD episodes'  
632 misclassifications can also be attributed to the lower accuracy of MODIS aerosol size retrievals over  
633 land (Section 2.1.1).

634

#### 635 4.1.2 $PM_{10}$ and dust contribution

636 The satellite algorithm's outputs, apart from AERONET retrievals, have been also compared  
637 against ground  $PM_{10}$  concentrations ( $\mu\text{g m}^{-3}$ ) measured in 22 Mediterranean stations (green triangles in  
638 Figure 1).

639 First, for each station, the number of intense DD episodes was calculated, for which coincident  
640 satellite and ground measurements (total  $PM_{10}$ ) are available (Figure 5-i). The number of concurrent  
641 DD episodes varies from 3 to 53, being in general decreasing from southern to northern stations. For 14  
642 out of 22 stations, where at least 10 intense DD episodes were identified by the satellite-based  
643 algorithm, we have computed the correlation coefficients between satellite  $AODs$  and surface total  
644  $PM_{10}$  concentrations (Fig. 5-ii). The highest  $R$  values (up to 0.8) are recorded in the central and eastern  
645 parts of the Mediterranean while the lowest ones are found in the western stations. It must be noted that  
646 the correlation coefficients are affected by outliers, because of the limited number of DD episodes in  
647 each station, highlighting the sensitiveness of the intercomparison. Such outliers can be expected when  
648 satellite-based columnar  $AODs$  and surface-based  $PM_{10}$  data are compared, since satellite  $AODs$  are  
649 representative for the whole atmospheric column in contrast to in-situ  $PM$  measurements which are  
650 more representative for the lowest part of the planetary boundary layer affected also by local factors.  
651 Therefore, the vertical distribution of desert dust load, as it will be presented in the next sections, can  
652 determine the level of agreement between satellite  $AODs$  and surface  $PM$  concentrations.

653 The identification method by the satellite algorithm can be considered as correct when dust  $PM_{10}$   
654 concentrations are higher than zero (i.e. dust has been recorded at the station). According to this, the  
655 ratio between the number of non-zero dust  $PM$  observations and the number of DD episodes  
656 (coincident satellite-derived DD episodes and total  $PM_{10}$  measurements) for each station is defined as  
657 success score. The calculated success scores (Figure 5-iii) vary from 68% (Monagrega, northeastern  
658 Spain, 28 episodes) to 97% (Boccadifalco, 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 (Figure 5-iv) is above 50%, ranging from 44% (Zarra, Spain) to 86.8% (Agia Marina, Cyprus). In order to complete our analysis we have also calculated the mean (Figure 5-v) and the median (Figure 5-vi) dust  $PM_{10}$  concentrations for the identified intense DD episodes in each station. The mean  $PM_{10}$  concentrations mainly vary between 20 and 50  $\mu\text{g m}^{-3}$ , being higher in the southern stations, as expected. The minimum mean value (17  $\mu\text{g m}^{-3}$ ) was recorded in Censt (Sardinia) and the maximum one (223  $\mu\text{g m}^{-3}$ ) in Agia 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  $PM_{10}$  during dusty days range from 8 to 23  $\mu\text{g m}^{-3}$  based on ground concentrations measured at 21 Mediterranean stations. These differences are reasonable since here only intense desert dust outbreaks associated with high aerosol optical depths are considered. Finally, the median  $PM_{10}$  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 Agia Marina (Cyprus), where the maximum daily  $PM_{10}$  concentrations, equal to 690 and 1291  $\mu\text{gm}^{-3}$ , respectively, were recorded during an intense dust outbreak that affected the eastern Mediterranean on 24 and 25 February 2006.

#### 4.2 2D geographical distributions of desert dust episodes' frequency and intensity

The mean geographical distributions of strong and extreme DD episodes' frequency of occurrence (episodes  $\text{yr}^{-1}$ ) are presented in Figure 6. Results are given separately as obtained from MODIS-Terra and Aqua for the periods Mar. 2000 – Feb. 2013 and 2003 – 2012, corresponding to local late morning-to-noon (Terra) and afternoon (Aqua) conditions, respectively. It is evident a gradual reduction of frequencies from South to North, while for the strong DD episodes also appears a West to East decreasing gradient. 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; 2014) 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. Marrioti 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 (Mar. 2000

690 – Feb. 2013). In general, there is similar spatial variability between Terra and Aqua, though slightly  
691 lower maximum frequencies are found for Aqua. Although intense dust episodes occur rarely across  
692 the northern parts of the study region ( $< 1$  and  $0.5$  episodes  $\text{yr}^{-1}$  for strong and extreme episodes,  
693 respectively), their occurrence proves that dust particles can be transported far away from their sources,  
694 up to the central (e.g. Klein et al., 2010) or even northern (e.g. Bègue et al., 2012) European areas  
695 under favorable meteorological conditions. Our calculated frequencies are significantly lower than the  
696 corresponding ones obtained in Pey et al. (2013), who studied the African dust intrusions towards the  
697 Mediterranean basin, based on ground *PM* concentrations, over the period 2001 – 2011. The observed  
698 deviations between the two studies are mainly attributed to the different thresholds definition and hence  
699 strength of dust episodes. Here, focus is given on the intense dust outbreaks (intensity equal/higher than  
700  $\text{Mean} + 2 * \text{Std}$ ) while in Pey et al. (2013) the dust occurrences were identified even at very low  
701 concentrations ( $> 1 \mu\text{g m}^{-3}$ ).

702 A noticeable difference between the two study periods and platforms is that relatively high  
703 frequencies of extreme DD episodes are recorded in more northern latitudes in the Mediterranean Sea,  
704 i.e. up to  $43^\circ$  N, according to MODIS-Terra over Mar. 2000 – Feb. 2013, while they are restricted  
705 south of  $40^\circ$  N parallel for MODIS-Aqua during 2003 – 2012. In order to investigate this difference in  
706 detail we have also applied the satellite algorithm, over the period 2003 – 2012, i.e. that of Aqua, using  
707 MODIS-Terra retrievals as inputs. Through this analysis (Figures S5 and S6 in the supplementary  
708 material), it is evident that there is a very good agreement between the satellite algorithm's outputs, for  
709 the periods Mar. 2000 – Feb. 2013 and 2003 – 2012, revealing a constant dust episodes' regime.  
710 Therefore, the discrepancy appeared between MODIS-Terra and MODIS-Aqua spatial distributions, is  
711 attributed to the diurnal variation of factors regulating the emission and transport of dust particles from  
712 the sources areas. Schepanski et al. (2009), analyzed the variation of the Saharan dust source activation  
713 throughout the day, based on MSG-SEVIRI satellite retrievals, reporting that dust mobilization is more  
714 intense in the local early morning hours after sunrise. Note, that desert dust episodes over the period  
715 Mar. 2000 – Feb. 2013 have been identified based on observations retrieved by the Terra satellite,  
716 which flies over the study region around noon in contrast to Aqua which provides aerosol  
717 measurements at early afternoon hours.

718 The analysis has been also repeated (results not shown here) considering as inputs to the satellite  
719 algorithm only *AODs* associated with cloud fractions lower/equal than 0.8, in order to investigate  
720 possible modifications to our results due to the cloud contamination effect. As it concerns the strong  
721 DD episodes, the geographical distributions are similar with those of Fig. 6, but the maximum

722 frequencies (recorded in Morocco) are higher by up to 2 episodes  $\text{yr}^{-1}$  and 0.3 episodes  $\text{yr}^{-1}$  for the  
723 MODIS-Terra (Mar. 2000 – Feb. 2013) and MODIS-Aqua (2003-2012) data set, respectively. On the  
724 contrary, in the case of extreme DD episodes the maximum frequencies decrease to 2.5 episodes  $\text{yr}^{-1}$   
725 for the period 2003 – 2012 and they shift southwards, namely over the northern coasts of Africa, while  
726 over the central parts of the Mediterranean Sea, they are lower than 1 episode  $\text{yr}^{-1}$ .

727 The maps of intensities (in terms of  $AOD_{550nm}$ ) of DD episodes (Figure 7), show that for both study  
728 periods and satellite platforms, the maximum intensities are over the Gulf of Sidra and the Libyan Sea,  
729 along the northern African coasts. These intensities reach  $AODs$  up to about 1.5 for strong and 4.1 for  
730 extreme episodes, while the minimum ones (values down to 0.25-0.46) are recorded in the northern and  
731 western Mediterranean parts. Note that dissimilar spatial patterns appear between the geographical  
732 distributions of DD episodes' frequency and intensity, indicating that these two features are determined  
733 by different factors (e.g. tracks or strength of depressions). Finally, when the cloud contamination is  
734 minimized using only  $AODs$  associated with  $CF$  lower than 0.8, then the maximum intensities are  
735 shifted southwards, across the northern Africa and eastern coasts of the Mediterranean, being lower  
736 than 1 and 2 for strong and extreme DD episodes, respectively. Through the rejection of possibly  
737 overestimated  $AODs$  from the dataset, it is found that the threshold levels are decreased (mainly over  
738 the most frequently dust affected areas) since both mean and standard deviation values are lower  
739 (results not shown here). Nevertheless, even though these  $AODs$  can be overestimated, in the majority  
740 of the cases the collocated AERONET  $AODs$  are high (but lower than the satellite observations)  
741 indicating the occurrence of desert dust outbreaks as it has also been shown in Section 4.1.1.4.

742 The analysis has been also repeated applying the alternative METHOD-B described in Section 3.  
743 Just to ensure a longer temporal coverage, this analysis was done for the period Mar. 2000 - Feb. 2013  
744 using MODIS-Terra data. The obtained results for the frequency of occurrence as well as for the  
745 intensity of DD episodes are depicted in Figures S7 and S8, respectively, in the supplementary  
746 material. The geographical patterns for the frequency of occurrence between the two methodologies are  
747 similar; however, the maximum frequencies of occurrence for the strong and extreme DD episodes can  
748 reach up to 13.3 episodes  $\text{yr}^{-1}$  (Fig. S7-i) and 8.1 episodes  $\text{yr}^{-1}$  (Fig. S7-ii), respectively. As it concerns  
749 the intensity, the geographical patterns, particularly for the strong DD episodes, are dissimilar and less  
750 distinct compared to the corresponding ones obtained with the primary methodology. This difference is  
751 attributed to the inclusion of more dust episodes with variable intensity, which leads to a not so clear  
752 “signal” when all these episodes are averaged. Based on METHOD-B, the maximum intensities (in  
753 terms of  $AOD_{550nm}$ ) of strong DD episodes can reach up to 1 (Fig. S8-i) while for the extreme episodes



754 (Fig. S8-ii) it can be as large as 3. The main finding, based on the intercomparison of the two  
755 methodologies for the identification of DD episodes, is that the frequency of the episodes is higher for  
756 the METHOD-B with respect to the primary methodology, while the intensity is decreased. Both facts  
757 are expected and can be explained by the lower calculated *AOD* thresholds with METHOD-B thus  
758 yielding more DD episodes of lower intensity.

759

#### 760 4.3 Vertical structure of the Mediterranean desert dust outbreaks

761 The ability of the developed satellite algorithm to detect intense dust episodes has been proved  
762 adequate through the comparison analysis against AERONET retrievals and *PM*<sub>10</sub> concentrations  
763 (Section 4.1). Nevertheless, its main limitation is that it uses columnar satellite retrievals and not  
764 vertical resolved data prohibiting thus the description of the vertical structure of these dust outbreaks.  
765 In order to address this issue, the CALIOP-CALIPSO retrievals are used as a complementary tool to  
766 the satellite algorithm's outputs. First, for the dust episodes identified by the satellite algorithm, the  
767 spatially and temporally collocated vertically resolved CALIOP lidar observations are selected. For  
768 these cases and for each 1° x 1° grid cell, we have divided the lower troposphere, up to 8 km, in 16  
769 layers of 500 meters height. In this way, 14400 boxes of 1° x 1° surface area and 500 meters height  
770 have been produced. Then, for each one of them, we have calculated the overall number of dust and  
771 polluted dust observations (hereafter named as dust) according to the aerosol subtyping scheme of the  
772 CALIOP Vertical Feature Mask (VFM). Note that dust and polluted dust were chosen because in  
773 previous studies (Mielonen et al., 2009) they were shown to be the best two defined aerosol types  
774 among the other ones classified by the CALIOP VFM. Nevertheless, in case of polluted dust, Burton et  
775 al. (2013) reported that dust particles can be mixed with marine aerosols instead of smoke or pollution  
776 as assumed by the VFM retrieval algorithm. In our study, more than 95% of the aerosol type records  
777 were pure dust, for the collocated cases between the satellite algorithm and CALIPSO observations. In  
778 addition, in the majority of the defined boxes, the percentage of dust from the overall observations is  
779 higher than 70%, confirming furthermore the validity of the algorithm DD episodes' identification  
780 procedure. This is an excellent proof of the successful identification of DD episodes by the satellite  
781 algorithm, since CALIOP-CALIPSO is an independent and vertically resolved platform and database.  
782 Thereby, CALIOP vertical observations were subsequently used to examine the vertical structure of  
783 dust outbreaks.

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

799 It should be noted that in the present work, we have analyzed all the available CALIOP overpasses  
800 (~ 10000) over the study region, during the period Jun. 2006 – Feb. 2013. For brevity reasons,  
801 however, only the obtained results based on MODIS-Terra retrievals are presented here, since similar  
802 findings are drawn for MODIS-Aqua (Jun. 2006 – Dec. 2012). Moreover, the analysis (results are not  
803 shown here) has been made separately for the identified strong and extreme DD episodes without  
804 revealing remarkable differences in the geometrical characteristics of dust outbreaks. Nevertheless, the  
805  $\beta_{532nm}$  values are higher for the extreme DD episodes being consistent with the discrimination of dust  
806 episodes' intensity (in terms of *AOD*), which is applied to the satellite algorithm. In order to facilitate  
807 the visualization of our results, for each column (1° x 1° spatial resolution) and latitudinal/longitudinal  
808 zone (1° degree), we have calculated the overall number of dust observations and the associated  
809 weighted averages of  $\beta_{532nm}$ , depending on the projection plane (latitudinal, longitudinal and columnar),  
810 according to dust observations in each box. For both parameters, the analysis has been made on an  
811 annual and seasonal basis and the corresponding results are discussed in Sections 4.3.1 and 4.3.2,  
812 respectively.

813

814

#### 815 4.3.1 Annual characteristics

816 In Figure 8, are presented the three dimensional structures of the CALIOP overall dust observations  
817 (Fig. 8-i) and the associated average total backscatter coefficients at 532 nm (Fig. 8-ii), during intense  
818 dust episodes conditions, over the broader Mediterranean area, for the period Jun. 2006 – Feb. 2013.  
819 From the latitudinal projection in Fig. 8-i, it is evident that dust particles are mainly detected between  
820 0.5 and 6 km, and more rarely up to 8 km, between the parallels 32° N and 38° N. The number of dust  
821 observations is increased at higher altitudes with increasing latitudes, up to 40° N, while the altitude  
822 range (thickness) where these records are detected is gradually reduced from 4 to 2 km. At northern  
823 latitudes, the CALIPSO dust records are drastically reduced and are mainly observed between 1 and 4  
824 km. The ascending mode of the transported mineral particles over the Mediterranean is attributed to the  
825 prevailing low pressure systems, which mobilize and uplift dust particles from the source areas across  
826 the Sahara Desert and the Arabian Peninsula. Dust aerosols are transported over the planetary boundary  
827 layer (Hamonou et al., 1999) due to the upward movement of dry and turbid air masses (Dulac et al.,  
828 1992), while the prevailing synoptic conditions determine also the spatial and temporal characteristics  
829 of desert dust outbreaks over the Mediterranean (Gkikas et al., 2015).

830 In general, our results are in agreement with previous studies, based on lidar profiles, which have  
831 been made in several Mediterranean sites. More specifically, Papayannis et al. (2008) found that dust  
832 layers, over the EARLINET Mediterranean stations, extend from 0.5 to 10 km above mean sea level,  
833 their center of mass is located between 2.5 and 3.5 km and their thickness ranges from 2.1 to 3.3 km.  
834 Hamonou et al. (1999) reported that dust layers are mainly detected between 1.5 and 5 km based on  
835 lidar measurements in the northwestern and northeastern Mediterranean. According to di Sarra et al.  
836 (2001), who studied the Saharan dust intrusions in Lampedusa (central Mediterranean) for the period  
837 May-June 1999, dust particles can be detected up to 7-8 km, which is in line with our findings for the  
838 corresponding latitudinal zones (35° N - 36 ° N). Balis (2012), analyzed 33 Raman/lidar profiles of  
839 Saharan dust intrusions over Thessaloniki (northern Greece), and found that the mean base and top of  
840 dust layers were equal to  $2.5\pm0.9$  and  $4.2\pm1.5$  km, respectively.

841 As to the variation of vertical extension with longitude (Fig. 8-i), it is revealed that the base height  
842 of dust layers is decreased towards the eastern parts of the study region. In the western Mediterranean,  
843 the mineral particles are mainly detected between 2 and 6 km while over the central and eastern  
844 Mediterranean the corresponding altitudes are equal to 0.5 and 6 km, respectively. It is well known,  
845 that dust is transported over the western Mediterranean mainly in summer (e.g. Moulin et al., 1998)

846 favored by low pressure systems located over the northwestern Africa (Gkikas et al., 2015) and the  
847 enhanced thermal convection, uplifting effectively dust aerosols at high altitudes in the troposphere.  
848 Moreover, air masses carrying dust particles are “convected” towards higher altitudes due to the  
849 existence of the Atlas Mountains Range. Therefore, the combination of strong convective processes  
850 over North Africa along with topography can explain the identification of dust aerosols at higher  
851 tropospheric levels over the western Mediterranean. It is the presence of mineral particles at high  
852 altitudes in western Mediterranean that can explain the poor-to-moderate agreement between  $PM_{10}$   
853 concentrations and MODIS  $AODs$  found in the Iberian Peninsula (Fig. 5-ii). In order to give a better  
854 insight to how the dust outbreaks’ vertical extension can affect the level of agreement between  
855 columnar  $AOD$  satellite retrievals and ground  $PM_{10}$  concentrations, emphasis is given at specific dust  
856 events and the relevant findings will be discussed in section 4.4. In the central and eastern parts of the  
857 Mediterranean basin, air masses carrying African dust aerosols travel at lower altitudes over Africa  
858 because of the absence of significant topographical objects on their route, as suggested by Pey et al.  
859 (2013).

860 Previous studies have shown that dust layers over the Mediterranean are characterized by a  
861 multilayered structure (e.g. Hamonou et al., 1999; Mona et al., 2006; Papayannis et al., 2008). This is  
862 also depicted in the longitudinal projection of Figure 8-i, where several dust layers of different base/top  
863 altitudes and geometrical thicknesses are detected. In general, the base heights vary from 0.5 to 2 km,  
864 the top heights from 4 to 6 km and the thicknesses from 1 to 4 km. The majority of common  
865 observations between the CALIOP profiles and the identified intense DD episodes by the satellite  
866 algorithm are recorded over the maritime parts of the study region (bottom map of Fig. 8-i). The  
867 maximum number of CALIOP dust observations ( $\sim 19000$ ) is recorded along the Atlantic coasts of  
868 Morocco, but high numbers (about 10000 – 15000) are also found across the northern African coasts.

869 Apart from the CALIOP dust observations, we have also analyzed the associated  $\beta_{532nm}$  values at  
870 the defined altitude ranges in order to describe the variation of intensity of the desert dust episodes with  
871 height over the Mediterranean (Fig. 8-ii). The maximum backscatter coefficients (up to  $0.006 \text{ km}^{-1} \text{ sr}^{-1}$ )  
872 are observed below 2 km, being increased towards the southern edges ( $30^\circ \text{ N} - 34^\circ \text{ N}$ ) of the study  
873 region, close to dust source areas. This is explained by the fact that dust particles due to their coarse  
874 size and large mass, are efficiently deposited and for this reason they are recorded at higher  
875 concentrations near to the source areas and at low altitudes. Nevertheless, the decreasing intensity with  
876 height towards the north is not so evident. Thus, high  $\beta_{532nm}$  values ( $\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) are observed  
877 between 2 and 4 km in the latitudinal zone extending from  $35^\circ \text{ N}$  to  $42^\circ \text{ N}$ . Though, the uppermost

altitudes where relatively high  $\beta_{532nm}$  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 (Figs 8-i and 8-ii) can be explained by the fact that  $\beta_{532nm}$  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  $\beta_{532nm}$  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  $\beta_{532nm}$  profiles (Fig. 8-ii) is less distinct compared to the corresponding one resulting from the latitudinal projection. Relatively high  $\beta_{532nm}$  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. 8-ii. It is reminded that higher intensities of dust episodes over the central and eastern Mediterranean have also been noticed based on MODIS retrievals (Figure 7). 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.

The three dimensional plots of Figures 8-i and 8-ii, have been also reproduced considering all the available dust and polluted dust CALIOP-CALIPSO records, without taking into account the satellite algorithm's outputs (intense dust outbreaks). The obtained results for the number of observations and  $\beta_{532nm}$  are presented in Figures S9-i and S9-ii, respectively. Note, that for each studied parameter the colorbar scales in Figure 8 and S9 are not identical because the number of observations for dust average conditions (Fig. S9-i) is extremely larger than the corresponding one during intense dust outbreaks (Fig. 8-i) while the opposite is found for the  $\beta_{532nm}$  values (Fig. 8-ii and Fig. S9-ii). It is apparent that the latitudinal projections calculated for the intense dust outbreaks (Fig. 8-i) and for all

the available CALIOP dust records (Fig. S9-i) reveal different patterns. More specifically, when all available CALIOP dust records are considered, it is found that dust aerosols are mainly confined between 1 and 3 km in the southernmost parts of the study region while the number of observations gradually decreases at higher altitudes and towards northern latitudes (Fig. S9-i). On the contrary, during dust outbreaks, mineral particles are transported over the Mediterranean following an ascending path, as it is depicted in the latitudinal projection of Figure 8-i. Nevertheless, it must be mentioned that over the desert areas there is a full coverage (see bottom map in Fig. S9-i) when all dust CALIOP records are considered in contrast to intense dust outbreaks (see bottom map in Fig. 8-i) attributed to the absence of DT retrievals, used as inputs to the satellite algorithm, over bright surfaces. The comparison between the longitudinal projections during intense dust outbreaks (Figure 8-i) and during average dust conditions (Fig. S9-i) reveals less remarkable differences than for the latitudinal projections. According to the longitudinal projection of Figure S9-i, in the western Mediterranean, dust layers are confined between 1 and 5 km, while their base and top altitude both decrease down to 0.5 and 4 km, respectively, for increasing longitudes. In the easternmost part of the study region, dust layers are mainly confined between 1 and 3 km, while its top height can reach up to 5 km. The intensity of dust loads (in terms of  $\beta_{532nm}$ ) is lower than  $0.003 \text{ km}^{-1} \text{ sr}^{-1}$  regardless the projection plane for average dust conditions based on CALIOP-CALIPSO lidar profiles (Fig. S9-ii). Moreover, the intensity of dust loads decreases gradually with height as well as from South to North revealing a distinct pattern in all projection planes in contrast to the corresponding ones found during desert dust outbreaks (Fig. 8-ii).

#### 4.3.2 Seasonal characteristics

The vertical structure of the Mediterranean desert dust outbreaks has also been analyzed separately for winter, spring, summer and autumn. The seasonal three dimensional representations of the CALIOP overall dust observations and the associated total backscatter coefficients are depicted in the left and right column of Figure 9, respectively. It must be noted, that  $\beta_{532nm}$  colorbars' ranges are common, among the seasons, depending on the projection plane. More specifically, the maximum limits have been set to  $0.012 \text{ km}^{-1} \text{ sr}^{-1}$ ,  $0.014 \text{ km}^{-1} \text{ sr}^{-1}$  and  $0.021 \text{ km}^{-1} \text{ sr}^{-1}$  for the latitudinal, longitudinal and bottom map projections, respectively. It should be mentioned that  $\beta_{532nm}$  values can reach up to  $0.045 \text{ km}^{-1} \text{ sr}^{-1}$ , but are associated with a very small number of dust observations.

939 The majority (85%) of dust observations is recorded in spring and summer, attributed to the  
 940 enhanced production rates of mineral particles and the prevailing atmospheric circulation over the  
 941 source areas and the Mediterranean. According to the latitudinal projections, a seasonal variability of  
 942 the intense Mediterranean desert dust outbreaks' geometrical characteristics is evident. Dust particles  
 943 are detected at higher altitudes (6-7 km) during warm seasons of the year while in winter they are  
 944 mainly detected below 3 km, and in autumn are recorded between 2 and 5 km. Nevertheless, it should  
 945 be mentioned that during these seasons only a small number of pixels (see bottom maps in Figs. 9 i-a,  
 946 iv-a) is available considering also that clouds prohibit the satellite observations. Note that in spring,  
 947 dust can be found at low tropospheric levels while in summer it is mainly observed above 1 km  
 948 highlighting thus the role of topography and the enhanced thermal convection. During the first half of  
 949 the year, the maximum dust observations are confined between the parallels 31° N and 37° N while  
 950 during the second one, they are shifted northwards in the latitudinal zone extending from 34° N to 40°  
 951 N. Similar latitudinal projections were also presented by Luo et al. (2015), for the same zonal areas of  
 952 the study region, who developed a new algorithm to improve CALIOP's ability to detect optically thin  
 953 dust layers. From the longitudinal projections as well as from the bottom maps, it is evident that the  
 954 maximum dust records are found in different Mediterranean sub-regions, depending on the season. The  
 955 geometrical characteristics, in longitudinal terms, of intense DD episodes affecting the western, central  
 956 and eastern parts of the Mediterranean are similar to those presented in the annual three dimensional  
 957 structure (Fig. 8-i) being more frequent in the eastern and central Mediterranean in winter, spring and  
 958 autumn and in the western and central Mediterranean in summer.

959 The seasonal patterns of  $\beta_{532nm}$  latitudinal projections are different than those for the dust  
 960 observations, while they also differ among the four seasons. The intensity of winter DD episodes is  
 961 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  
 962 the longitudinal and bottom map projections, these episodes take place over the central and eastern  
 963 Mediterranean Sea but the number of grid cells with coincident CALIOP observations and DD episodes  
 964 is limited. In spring, the highest  $\beta_{532nm}$  values (up to  $0.006 \text{ km}^{-1} \text{ sr}^{-1}$ ) are recorded between the parallels  
 965 31° N and 35° N and below 2 km, although, relatively high  $\beta_{532nm}$  values (up to  $0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) are  
 966 found up to 5 km (Fig. 9 ii-b). Moving northwards, over the Mediterranean, dust layers are mainly  
 967 confined between 2 and 4 km, associated with high  $\beta_{532nm}$  values (up to  $0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) in the  
 968 latitudinal zone extending from 35° N to 43° N. The existence of these elevated dust layers, has been  
 969 also confirmed by model simulations through specific (Papayannis et al., 2008; 2014) or averaged  
 970 (Alpert et al., 2004) cross sections of dust concentrations in the central sector of the Mediterranean.

971 This is in accordance with our longitudinal projection (Fig. 9 ii-b), where  $\beta_{532nm}$  is high varying from  
972 0.004 to 0.008 km<sup>-1</sup> sr<sup>-1</sup> at these altitude ranges.

973 In summer, the intensity of dust episodes is smoothly decreased at higher altitudes, where dust  
974 layers of considerable  $\beta_{532nm}$  values are also found. More specifically, the highest backscatter  
975 coefficients (up to 0.008 km<sup>-1</sup> sr<sup>-1</sup>) are recorded near to the surface but also moderate values (up to  
976 0.006 km<sup>-1</sup> sr<sup>-1</sup>) are observed between 2 and 5 km, particularly over the southern parts of the study  
977 region (Fig. 9 iii-b). Most of these intense DD episodes occur in the western Mediterranean, where the  
978 highest  $\beta_{532nm}$  values (up to 0.005 km<sup>-1</sup> sr<sup>-1</sup>) are recorded between 2 and 5 km. Over the central and  
979 eastern Mediterranean, even higher  $\beta_{532nm}$  values are found (up to 0.014 km<sup>-1</sup> sr<sup>-1</sup>) but at lower altitudes  
980 (< 1 km). In autumn, the majority of the grid cells of coincident CALIOP profiles and DD episodes  
981 identified by the satellite algorithm are located between the parallels 33° N and 41° N. In this  
982 latitudinal zone, CALIOP profiles are available over the interior parts of the Iberian Peninsula and over  
983 western and central parts of the Mediterranean Sea, near to the northern African coasts. According to  
984 the latitudinal projection,  $\beta_{532nm}$  values mainly vary from 0.002 to 0.009 km<sup>-1</sup> sr<sup>-1</sup>, revealing an  
985 increasing tendency for increasing heights. On the contrary, the total backscatter coefficients do not  
986 show a distinct spatial pattern on the longitudinal projection, due to the limited number of grid cells  
987 participating in the calculations. Throughout the year, based on the CALIOP  $\beta_{532nm}$  retrievals, the DD  
988 episodes are more intense (up to 0.018 km<sup>-1</sup> sr<sup>-1</sup>) in spring, when massive dust loads are transported  
989 from the Sahara desert towards the central and eastern parts of the Mediterranean Sea (bottom map in  
990 Fig. 9 ii-b).

991

#### 992 4.4. Intercomparison of satellite AOD and PM<sub>10</sub> concentrations for specific desert dust outbreaks

993 In Section 4.1.2, it has been shown that the agreement between the satellite algorithm's outputs and  
994 PM<sub>10</sub> concentrations is better in the central and eastern Mediterranean with regards to the western parts  
995 (Figure 5-ii). This discrepancy has been mainly attributed to the higher altitude of dust layers' base  
996 over the western sector of the study domain (Figure 8-i), in relation to the existing areal orography.  
997 Here, aiming at addressing how dust layers' geometrical characteristics influence the agreement  
998 between columnar AOD satellite and ground PM<sub>10</sub> measurements, specific desert dust outbreaks that  
999 took place over the PM<sub>10</sub> stations are analyzed. These outbreaks were selected based on concurrent  
1000 fulfillment of the following criteria: (i) a DD episode must be identified by the satellite algorithm at  
1001 pixel level (at 1° x 1° grid cell), (ii) total PM<sub>10</sub> measurement must be available at the station which lies



into the geographical limits of the corresponding grid cell and (iii) CALIPSO flies across the grid cell. These criteria were met for 13 desert dust outbreaks, which took place over 9  $PM_{10}$  stations during the period Jun. 2006 – Feb. 2013. Similarities were found among the identified cases and therefore only the results for four desert dust outbreaks of different geometrical characteristics are discussed in the present section. For each case, we have reproduced the cross sections of the  $\beta_{532nm}$  vertical profiles up to 8 km above sea level (a.s.l.) along the CALIOP-CALIPSO track when the satellite flies near the  $PM_{10}$  site (Figures 10-12). Moreover, the corresponding aerosol subtype profiles, acquired from the CALIOP website ([http://www-calipso.larc.nasa.gov/products/lidar/browse\\_images/production/](http://www-calipso.larc.nasa.gov/products/lidar/browse_images/production/)), are provided in the supplementary material (Figures S10-S12). Since the  $PM_{10}$  concentrations are available only as daily averages, the optimum solution would be to have the maximum number (2) of CALIOP overpasses near  $PM_{10}$  site throughout the day, in order to reduce the temporal inconsistencies between satellite vertical resolved retrievals and ground data. However, in 8 out of 13 desert dust outbreaks this was not feasible.

1015

#### 1016 4.4.1 Case 1: 26<sup>th</sup> May 2008

1017 The first study case refers to a desert dust outbreak that took place on 26<sup>th</sup> May 2008 and affected  
 1018 the station Censt (Lat: 39.064, Lon: 8.457) located in southern Sardinia. At the ground, the measured  
 1019 mean daily total  $PM_{10}$  concentration was  $19 \mu\text{g m}^{-3}$  whereas 68% (or  $13 \mu\text{g m}^{-3}$ ) of the load consisted of  
 1020 dust particles indicating thus their strong presence in the lowest troposphere. Based on MODIS-Terra  
 1021 retrievals, representative for the whole atmospheric column and grid cell, the aerosol optical depth at  
 1022 550 nm was equal to 0.81. In order to investigate the vertical distribution of the dust outbreak, the cross  
 1023 sections of the  $\beta_{532nm}$  vertical profiles along CALIOP track, near the station, during daytime and  
 1024 nighttime have been reproduced and depicted in Figures 10-i and 10-ii, respectively. In addition, the  
 1025 corresponding aerosol subtype profiles are provided in Figures S10-i and S10-ii in the supplementary  
 1026 material. During night, the predominance of a well-developed dust layer mixed with polluted aerosols  
 1027 is evident (Figure S10-i) extending from surface up to 5 km a.s.l. between the parallels 33° N and 38°  
 1028 N, while near the station its top is lowered down to 3 km (left side of Figure 10-i). Moreover, the  $\beta_{532nm}$   
 1029 values range mainly from 0.002 to  $0.003 \text{ km}^{-1} \text{ sr}^{-1}$  without revealing remarkable variations, thus  
 1030 indicating a rather compact dust layer. According to the daytime CALIOP overpass (Figure 10-ii), a  
 1031 pure dust layer (Figure S10-ii) is confined between surface and 4 km, affecting the surrounding area of  
 1032 the station, while its intensity (in terms of  $\beta_{532nm}$ ) varies slightly from 0.0015 to  $0.002 \text{ km}^{-1} \text{ sr}^{-1}$ .

1033 Nevertheless, due to the background solar illumination, leading thus to a lower signal-to-noise ratio  
1034 (Nowottnick et al., 2015), the “borders” of the dust plume during daytime are not so distinct in contrast  
1035 to nighttime. According to the obtained results, the ground-based measurements are able to capture  
1036 satisfactorily the dust event when its load is equally distributed in the lowest tropospheric levels,  
1037 resulting thus to a good agreement between MODIS and  $PM_{10}$  observations.

1038

#### 1039 4.4.2 Case 2 and 3: 16<sup>th</sup> July 2008 and 12<sup>th</sup> September 2007

1040 Two dust events that affected Els Torms (NE Spain, Lat: 41.395, Lon: 0.721) and San Pablo  
1041 (central Spain, Lat: 39.525, Lon: -4.353) on 16<sup>th</sup> July 2008 and 12<sup>th</sup> September 2007, respectively, are  
1042 studied here. The daily averages of the total  $PM_{10}$  concentrations were equal to 16 and 30  $\mu\text{g m}^{-3}$ ,  
1043 respectively, whereas the dust particles’ contribution (dust  $PM_{10}$ ) to the total amount was zero in Els  
1044 Torms and 33 % in San Pablo. On the contrary, the MODIS-Terra level 3  $AOD$  retrievals were high and  
1045 equal to 0.56 (Els Torms) and 0.64 (San Pablo), indicating the existence of dust aerosols according to  
1046 the satellite algorithm’s classification method. In order to give a better insight, aiming at describing the  
1047 discrepancies between MODIS-Terra  $AOD$  and  $PM_{10}$  concentrations, we have reproduced the cross  
1048 sections of the total backscatter at 532 nm when CALIPSO flies, during daytime, near Els Torms  
1049 (Figure 11-i) and San Pablo (Figure 11-ii). The corresponding profiles of the CALIOP aerosol  
1050 classification scheme are also available in Figures S11-i and S11-ii. In Els Torms, where the dust  $PM_{10}$   
1051 concentration was zero, a dust layer (Figure S11-i) with its base at 3.5 km a.s.l. and its top at 5 km  
1052 a.s.l., is recorded by the CALIOP lidar between the parallels 41° N and 43° N. The intensity of the  
1053 elevated dust layer, in terms of  $\beta_{532nm}$ , varies from 0.002 to 0.004  $\text{km}^{-1} \text{sr}^{-1}$  (Figure 11-i). Through  
1054 CALIOP lidar profiles, it is confirmed the existence of a dust layer aloft, which cannot be captured by  
1055 the  $PM_{10}$  measurements in contrast to the MODIS spectroradiometer. In San Pablo, where the dust  
1056 particles’ contribution to the total  $PM_{10}$  load was equal to 33 %, a dust layer abuts the ground extending  
1057 up to 5-6 km a.s.l., whereas the dust plume covers a wide range, in latitudinal terms, from the sub-Sahel  
1058 to the Celtic Sea, affecting the Iberian Peninsula (Figure S11-ii). Nevertheless, the intensity of the dust  
1059 layer, over the surrounding area of the station, differs with altitude being higher between 2.5 and 5 km  
1060 a.s.l. (0.004 to 0.007  $\text{km}^{-1} \text{sr}^{-1}$ ) and lower between ground and 2 km a.s.l. ( $< 0.003 \text{ km}^{-1} \text{sr}^{-1}$ ), as  
1061 depicted in the middle of Figure 11-ii. The two studied cases here differ from Case 1 (Section 4.4.1)  
1062 either with regards to the position of the elevated dust layer (Els Torms) or to its vertical distribution

1063 (San Pablo), which explains the poor agreement between satellite columnar *AOD* retrievals (MODIS)  
1064 and ground *PM*<sub>10</sub> concentrations.

1065

#### 1066 4.4.3 Case 4: 25<sup>th</sup> February 2007

1067 The case studied here, namely the desert dust outbreak recorded in Agia Marina (Cyprus, Lat:  
1068 35.039, Lon: 33.058) on 25<sup>th</sup> February 2007, is the strongest one among the selected cases. More  
1069 specifically, the daily average of the dust *PM*<sub>10</sub> concentration was equal to 134 µg m<sup>-3</sup> accounting for  
1070 the 92 % of the total *PM*<sub>10</sub> measured amount at the station, which is indicative of the strong  
1071 predominance of dust particles in the lowest troposphere. The MODIS-Terra level 3 *AOD* value for the  
1072 grid cell to which the station belongs to is high and equal to 1.04. According to the CALIOP aerosol  
1073 classification scheme, during nighttime, a shallow low-elevated dust layer mixed with polluted or  
1074 marine aerosols is heading towards the station, whereas above the *PM*<sub>10</sub> site (Agia Marina) it extends  
1075 from close to the ground up to 9 km a.s.l., comprising mainly pure dust aerosols (Figure S12). The  
1076 main part of the dust layer, in the surrounding area of the station, is confined between 2.5 and 4 km  
1077 a.s.l. where the maximum  $\beta_{532nm}$  values (up to 0.006 km<sup>-1</sup> sr<sup>-1</sup>) are observed (Figure 12). Also, similar  
1078  $\beta_{532nm}$  values are recorded below 1 km a.s.l.; however, the dust layer is not well represented in the cross  
1079 section of the CALIOP  $\beta_{532nm}$  vertical profiles due to the total attenuation of the lidar beam by clouds  
1080 (located between 3 and 4 km a.s.l.) superimposed to the low-elevated dust layer.

1081

## 1082 5. Summary and conclusions

1083 This study aims at describing the vertical structure of intense desert dust outbreaks affecting the  
1084 broader Mediterranean basin. To achieve this target, an updated version of an objective and dynamic  
1085 algorithm, which has been introduced by Gkikas et al. (2009; 2013), has been applied for the  
1086 identification of strong and extreme desert dust episodes, over the period Mar. 2000 – Feb. 2013. For  
1087 its operation, a group of optical properties, retrieved by satellite sensors (MODIS-Terra/Aqua, EP-  
1088 TOMS and OMI-Aura) on a daily basis, is used, providing information about aerosols' load, size and  
1089 nature. Briefly, the satellite algorithm consists of three steps; at the first one are computed the mean  
1090 *AOD* value (*Mean*) and the associated standard deviation (*Std*) for the whole study period in each grid  
1091 cell of 1° x 1° spatial resolution, at the second one the identified aerosol episodes are classified based  
1092 on their intensity into strong and extreme ones. Finally, at the third step the desert dust episodes are

1093 identified among these, separately over land and sea. Through this approach the selected dataset  
1094 consists only of intense desert dust episodes since their intensity (expressed in terms of  $AOD_{550nm}$ ) is  
1095 higher/equal than/to  $Mean + 2*Std$ . The DD episodes have also been determined by applying an  
1096 alternative second methodology (METHOD-B), which excludes dust-affected cases identified based on  
1097 the criteria set concerning the aerosol size/nature related optical properties.

1098 Through the comparison of the default version of the satellite algorithm against surface  
1099 measurements derived from 109 AERONET and 22  $PM_{10}$  stations, it is found that:

#### 1100 AERONET

- 1101 ➤ The correlation coefficient between MODIS and AERONET  $AODs$  is increased from 0.505 to  
1102 0.750 when level 3 grid cells with higher sub-grid spatial representativeness and homogeneity  
1103 are considered.
- 1104 ➤ According to the AERONET volume size distributions, the predominance of the coarse mode is  
1105 evident with a peak ( $\sim 0.25 \mu m^3 \mu m^{-2}$ ) for particles radii between 1.70 and 2.24  $\mu m$ , in case of  
1106 intense DD episodes.
- 1107 ➤ The appropriateness of DD episodes' identification method applied to the satellite algorithm is  
1108 confirmed since the majority ( $> 75\%$ ) of AERONET  $\alpha_{440-870nm}$  and  $r_{eff}$  values are lower than  
1109 0.54 and higher than 0.55  $\mu m$ , respectively.
- 1110 ➤ About 15% of the pixel level intense DD episodes are misclassified by the satellite algorithm  
1111 and these drawbacks are encountered in AERONET stations where the aerosol load is  
1112 dominated either by fine particles or by complex aerosol types.

#### 1113 $PM_{10}$ and dust contribution

- 1114 ➤ The agreement between surface and satellite measurements is better over the central and eastern  
1115 Mediterranean stations.
- 1116 ➤ On a station level, the percentage of the intense DD episodes, for which a dust contribution to  
1117  $PM_{10}$  surface concentration has been recorded, varies from 68% (Monagrega, northeastern  
1118 Spain) to 97% (Boccadifalco, Sicily).
- 1119 ➤ In the majority of stations, dust particles contribute more than 50% of the total amount reaching  
1120 up to 86.8% (Agia Marina, Cyprus).
- 1121 ➤ The mean  $PM_{10}$  concentration levels mainly vary from 20 to 50  $\mu g m^{-3}$  reaching up to 223  $\mu g$   
1122  $m^{-3}$  in Agia Marina (Cyprus).

1123 Based on the satellite algorithm's outputs, an overall view about the regime of Mediterranean desert  
1124 dust outbreaks is presented for the periods Mar. 2000 – Feb. 2013 (MODIS-Terra) and 2003-2012  
1125 (MODIS-Aqua). The main findings concerning the intense DD episodes' frequency (in terms of  
1126 episodes yr<sup>-1</sup>) and intensity (in terms of *AOD* at 550nm) are the following:

- 1127 ➤ Strong DD episodes occur more frequently (up to 9.9 episodes yr<sup>-1</sup>) in the western  
1128 Mediterranean while the extreme ones occur more frequently (up to 3.3 episodes yr<sup>-1</sup>) over the  
1129 central parts of the Mediterranean Sea, when the satellite algorithm operates with MODIS-Terra  
1130 retrievals.
- 1131 ➤ The intensity of strong and extreme DD episodes, in *AOD* terms, can reach to 1.5 and 3-4,  
1132 respectively, over the central and eastern parts of the Mediterranean Sea, near off the northern  
1133 African coasts.
- 1134 ➤ Slightly lower frequencies and higher intensities are found for the period 2003-2012, when the  
1135 satellite algorithm operates with MODIS-Aqua retrievals.
- 1136 ➤ Through the intercomparison between the two applied methodologies, it is revealed that the  
1137 geographical patterns of frequency of occurrence are similar both for strong and extreme DD  
1138 episodes; however, higher frequencies are found based on METHOD-B.
- 1139 ➤ Based on METHOD-B, the DD episodes' intensities are decreased whereas the geographical  
1140 patterns for the strong DD episodes are not so distinct compared to the corresponding results  
1141 obtained by the default version of the satellite algorithm.
- 1142 ➤ The similarity between the outputs of the algorithm using the two methodologies shows the  
1143 consistency of the algorithm and the validity of its concept.

1144  
1145 In order to describe the vertical structure of the intense Mediterranean dust outbreaks, the CALIOP  
1146 vertical profiles of aerosol subtyping and total backscatter coefficient at 532 nm, are used as a  
1147 complementary tool to the identified intense DD episodes by the satellite algorithm. Through this  
1148 synergistic approach it is found that:

- 1149 ➤ Dust particles are mainly detected between 0.5 and 6 km, following an ascending mode, up to  
1150 40° N, leaving from the source areas and transported towards the Mediterranean.
- 1151 ➤ Over the western Mediterranean, the dust layers are mainly observed between 2 and 6 km while  
1152 their base height is decreased down to 0.5 km for increasing longitudes.
- 1153 ➤ During the warm period of the year, dust particles are uplifted at higher altitudes (up to 8 km).

- In summer, the transported dust loads over the western Mediterranean are recorded above 1 km and in spring at lower altitudes over the central and eastern parts of the study region. This behavior underlies the role of topography (e.g. Atlas Mountains) and the enhanced thermal convection.
- The intensity of dust outbreaks, in terms of  $\beta_{532nm}$ , is maximized (up to  $0.006 \text{ km}^{-1} \text{ sr}^{-1}$ ) below 2 km and at the southern parts ( $30^\circ \text{ N} - 34^\circ \text{ N}$ ) of the study region.
- In spring, considerably high  $\beta_{532nm}$  values ( $\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) are observed between 2 and 4 km in the latitudinal zone extending from  $35^\circ \text{ N}$  to  $42^\circ \text{ N}$ .
- Moderate-to-high  $\beta_{532nm}$  values are observed up to 6 km, near to the source areas, while the top of dust layers is gradually decreased down to 4 km towards northern latitudes.
- From the longitudinal projection of  $\beta_{532nm}$ , it is evident that DD episodes are more intense ( $\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$ ) between 1 and 5 km in the western Mediterranean, while over the central and eastern sectors, the maximum intensities ( $\sim 0.006 \text{ km}^{-1} \text{ sr}^{-1}$ ) are recorded below 1.5 km.
- On a seasonal basis, DD episodes are found to be more intense (up to  $0.018 \text{ km}^{-1} \text{ sr}^{-1}$ ) in spring, when dust is transported towards the central and eastern parts of the Mediterranean region.

At the last part of the present study, it is investigated how the desert dust outbreaks' vertical distribution can affect the level of agreement between columnar satellite *AOD* retrievals (MODIS) and ground *PM*<sub>10</sub> concentrations. For this purpose, four intense Mediterranean desert dust outbreaks of different geometrical characteristics that took place across the Mediterranean, namely in Spain (western), Italy (central) and Cyprus (eastern), are studied when satellite algorithm's outputs, ground *PM*<sub>10</sub> concentrations and CALIOP-CALIPSO lidar profiles are available concurrently. Our analysis clearly shows that when a well-developed and compact dust layer is located in the lowest tropospheric levels, then the level of agreement between MODIS-*PM*<sub>10</sub> is high. On the contrary, when the dust layer is aloft or its load is not equally distributed in vertical, then a poor agreement between MODIS-*PM*<sub>10</sub> is found.

This study attempts to highlight the importance of the synergistic use of active and passive satellite observations and the usage of surface-based measurements, targeting to the representation of the 3D structure of dust outbreaks and the description of their spatial and temporal features. For this reason, the further development of the satellite algorithm is an ongoing process by our group, aiming at extending the study domain from regional to global scale, considering the latest version of MODIS retrievals (Collection 006) as well as the Deep Blue Algorithm retrievals, available over the major dust sources of the planet.

1186

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1207

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1705 **Table 1:** AERONET stations, depicted with cyan colors in Figure 1, used for the identification of desert dust (DD) episodes  
 1706 based on ground retrievals.

Stations	Latitude	Longitude	Study period
<b>Blida</b>	N 36° 30' 28"	E 02° 52' 51"	7 Nov. 2003 – 18 Feb. 2012
<b>El Arenosillo</b>	N 37° 06' 18"	W 06° 43' 58"	1 Mar. 2000 – 21 Feb. 2010
<b>Evora</b>	N 38° 34' 04"	W 07° 54' 43"	4 Jul. 2003 – 28 Feb. 2013
<b>FORTH CRETE</b>	N 35° 19' 58"	E 25° 16' 55"	23 Jan. 2003 – 6 Aug. 2011
<b>IMC Oristano</b>	N 39° 54' 36"	E 08° 30' 00"	30 May 2000 – 28 Feb. 2003
<b>IMS METU Erdemli</b>	N 36° 33' 54"	E 34° 15' 18"	1 Mar. 2000 – 28 Feb. 2013
<b>Nes Ziona</b>	N 31° 55' 19"	E 34° 47' 20"	1 Feb. 2000 – 28 Feb. 2013

1707

1708 **Table 2:** Percentages of the satellite Ångström exponent, Fine fraction, Effective Radius and Aerosol Index retrievals  
 1709 satisfying the defined thresholds in the satellite algorithm for the identification of desert dust episodes.

Parameter	Valid	Invalid	Number of DD episodes
<b>Ångström exponent</b>	97.8%	2.2%	232
<b>Fine fraction</b>	98.7%	1.3%	232
<b>Effective radius</b>	94.5%	5.5%	117
<b>Aerosol Index</b>	86.9%	13.1%	206

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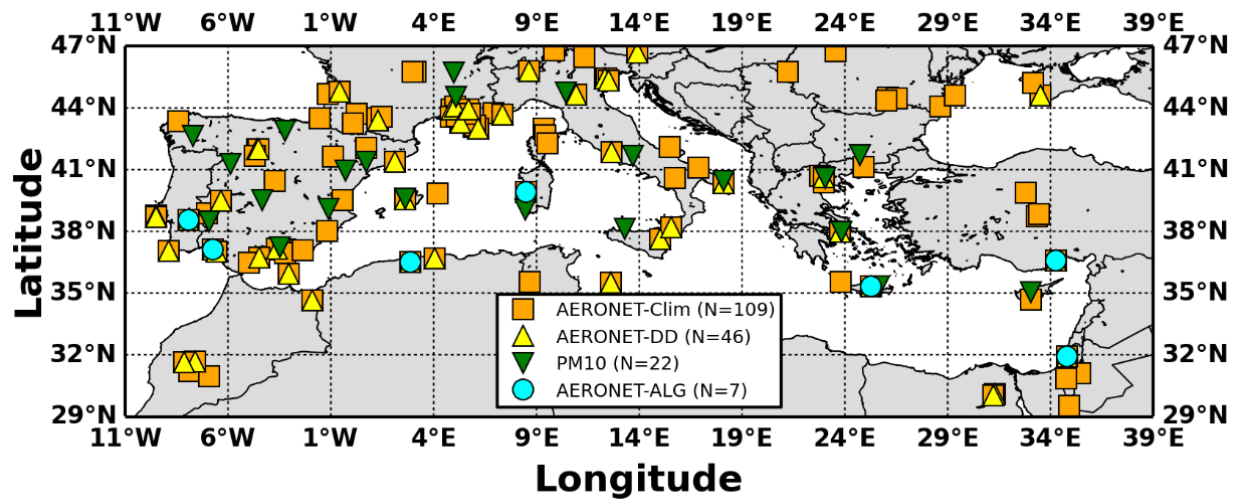
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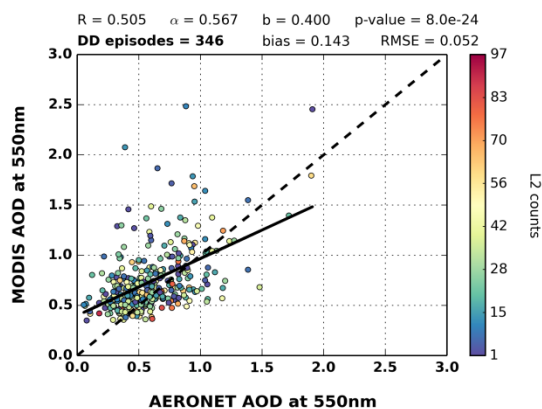
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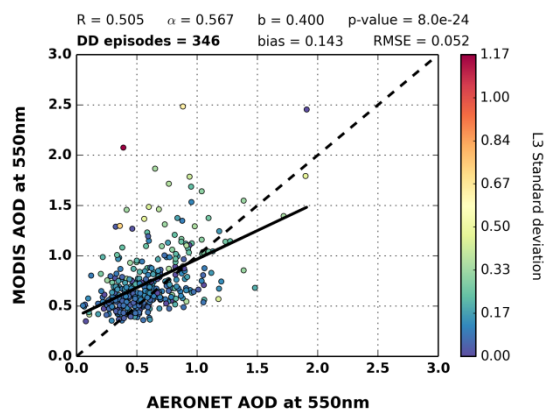
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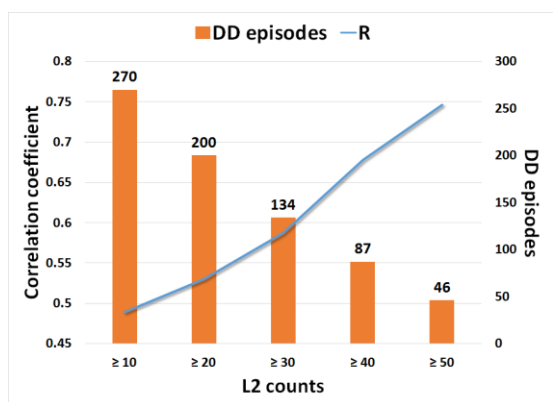
**Figure 1:** Locations of the AERONET and  $PM_{10}$  stations that have been used for the evaluation of the algorithm's outputs. More specifically, with orange squares are denoted the AERONET stations located into the study region, with the yellow triangles, the AERONET stations with coincident satellite and ground retrievals under dust episodes conditions, with the cyan circles, the AERONET stations which have been used for the evaluation of the defined algorithm thresholds, and with the green triangles are depicted the  $PM_{10}$  stations.



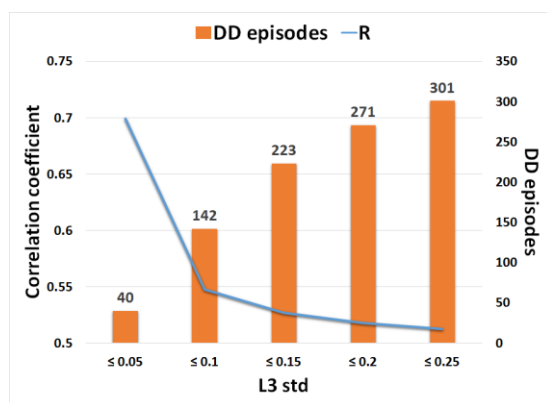
(i-a)



(i-b)

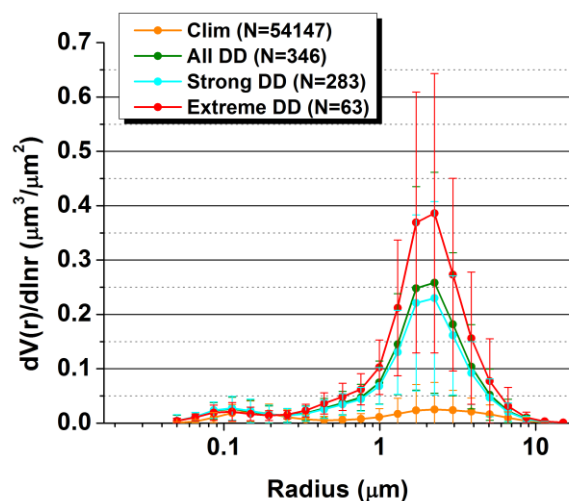


(ii-a)

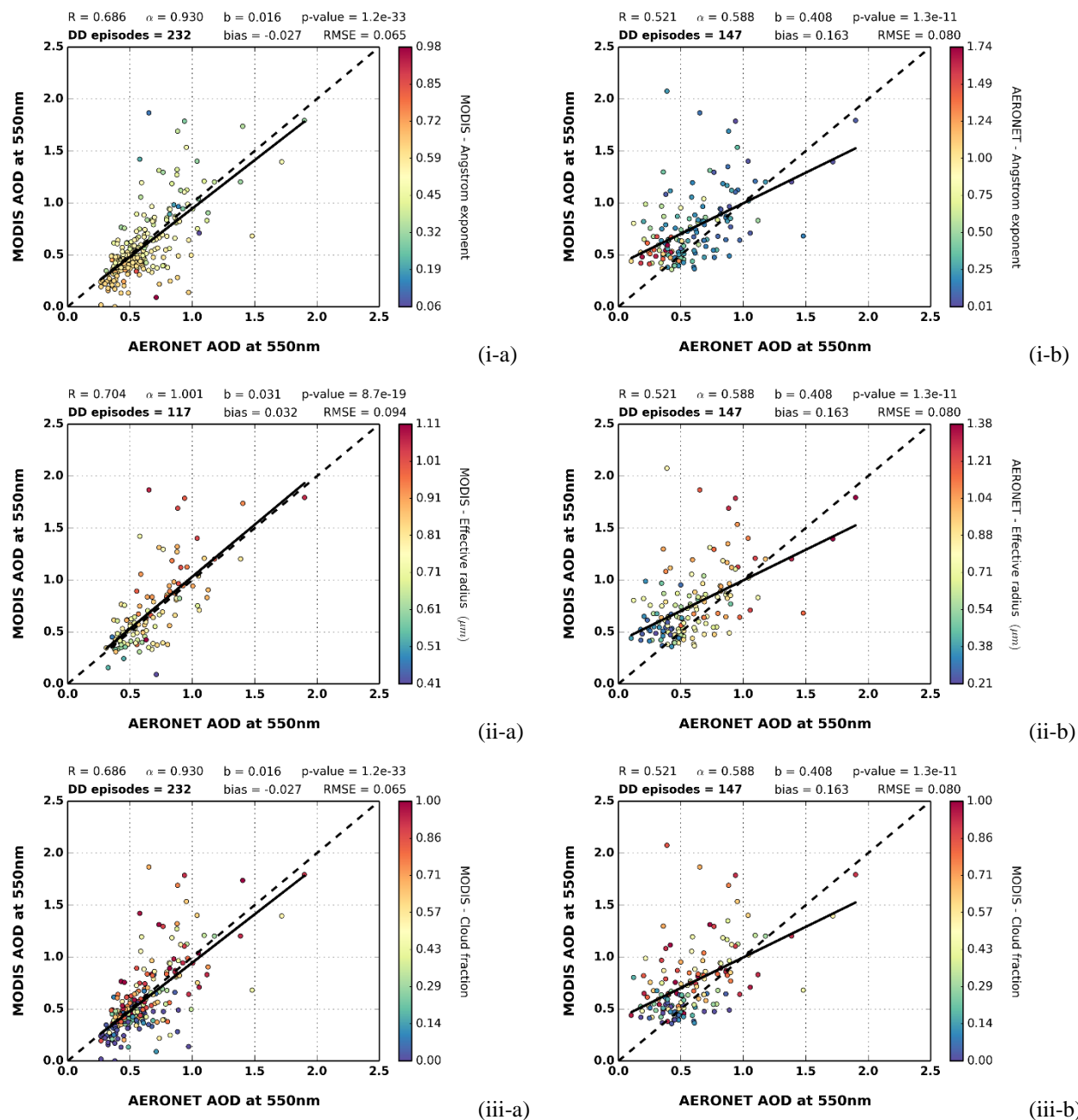


(ii-b)

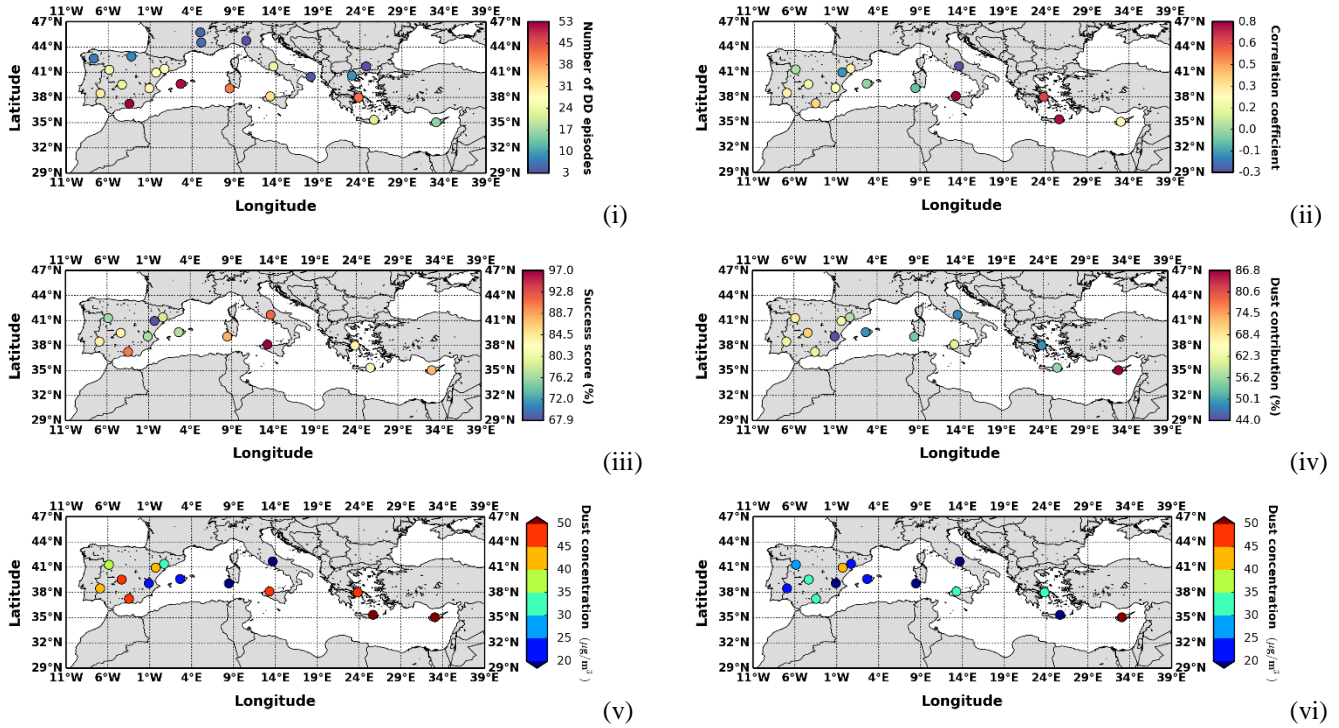
**Figure 2:** (i) Scatterplots between MODIS-Terra and AERONET aerosol optical depths at 550 nm under intense desert dust episodes conditions related to the: (a) number of level-2 counts which are used for the calculation of the level-3 retrievals and (b) spatial standard deviation inside the  $1^\circ \times 1^\circ$  grid cells (level-3 retrievals). (ii) Sensitivity analysis for the calculated correlation coefficients between satellite and ground *AODs*, depending on the: (a) number of level-2 retrievals and (b) sub-grid standard deviation of level-3 retrievals.



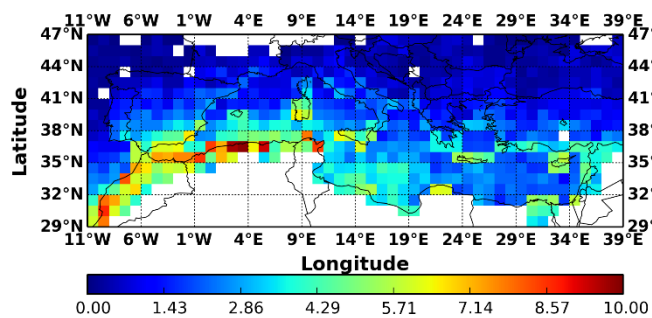
**Figure 3:** AERONET size distributions averaged over all available retrievals (orange curve) as well as over the total (green curve), strong (cyan curve) and extreme (red curve) desert dust episodes that occurred over the broader area of the Mediterranean basin, during the period Mar. 2000 – Feb. 2013. The error bars represent the calculated standard deviations.



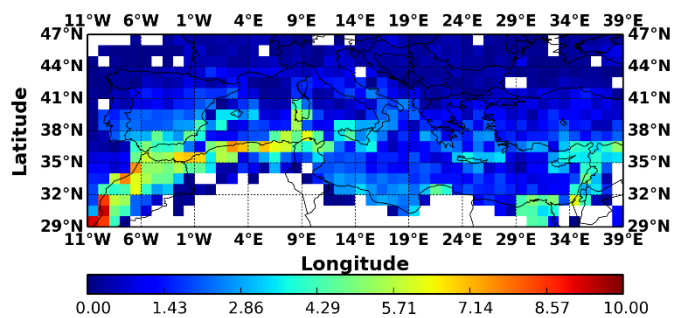
**Figure 4:** Scatterplots of MODIS-Terra and AERONET aerosol optical depths at 550 nm when intense dust episodes have been identified based on: (a) AERONET retrievals and (b) satellite algorithm, respectively. In the left column, colormaps indicate the corresponding values of: (i) Ångström exponent, (ii) Effective radius and (iii) Day cloud fraction derived by MODIS-Terra retrievals. In the right column, colormaps indicate the corresponding values of: (i) AERONET Ångström exponent, (ii) AERONET Effective radius and (iii) MODIS day cloud fraction retrievals. For each scatterplot, are provided the correlation coefficient ( $R$ ), slope ( $\alpha$ ), intercept ( $b$ ), p-value, number of DD episodes, bias (MODIS – AERONET) and root mean square error ( $RMSE$ ).



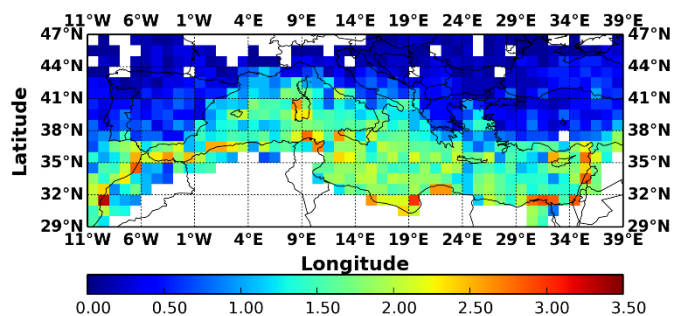
**Figure 5:** (i) Number of concurrent intense DD episodes where total  $PM_{10}$  concentrations and MODIS-Terra  $AOD$  retrievals are available, (ii) Computed correlation coefficient values between total  $PM_{10}$  concentrations and MODIS-Terra  $AOD$  retrievals in stations where at least 10 DD episodes have been recorded, (iii) Percentage of intense DD episodes where dust particles have been identified by the ground stations, (iv) Dust contribution percentages (%) to the total  $PM_{10}$  concentrations, (v) Calculated mean and (vi) median dust concentrations ( $\mu\text{g m}^{-3}$ ), based on ground measurements for the identified intense DD episodes by the satellite algorithm.



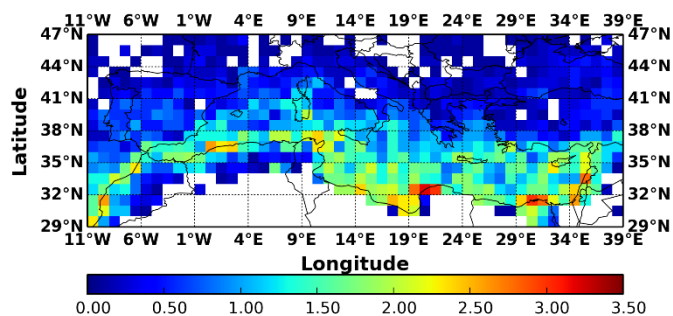
(i-a)



(i-b)

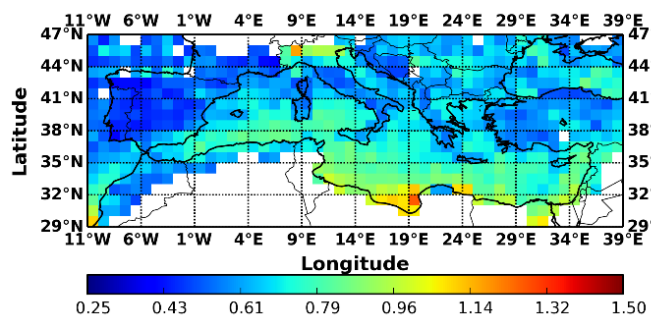


(ii-a)

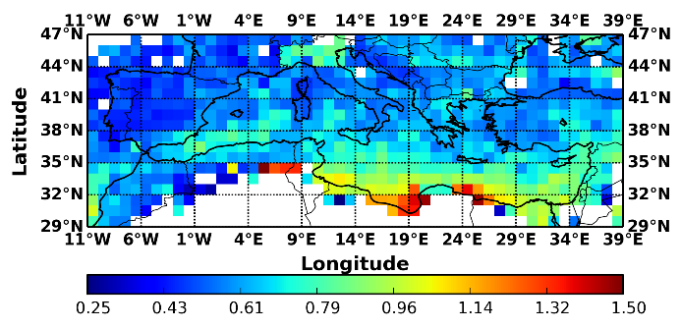


(ii-b)

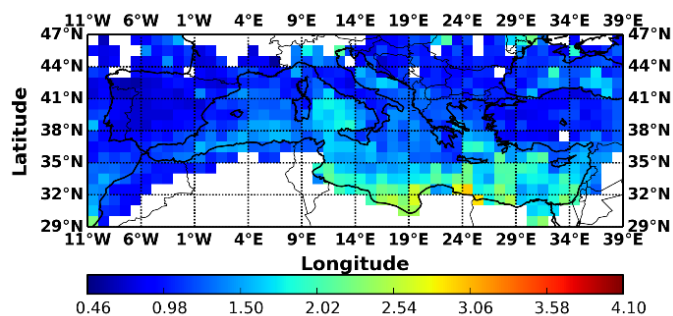
**Figure 6:** Geographical distributions of the occurrence frequency (episodes  $\text{yr}^{-1}$ ) of: (i) strong and (ii) extreme desert dust episodes, averaged over the periods: (a) Mar. 2000 – Feb. 2013 (MODIS-Terra) and (b) 2003 – 2012 (MODIS-Aqua), over the broader area of the Mediterranean basin.



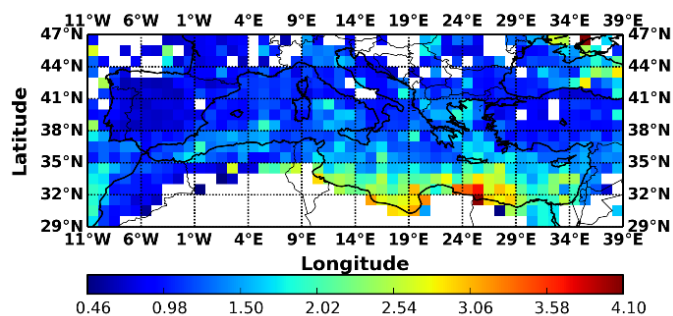
(i-a)



(i-b)



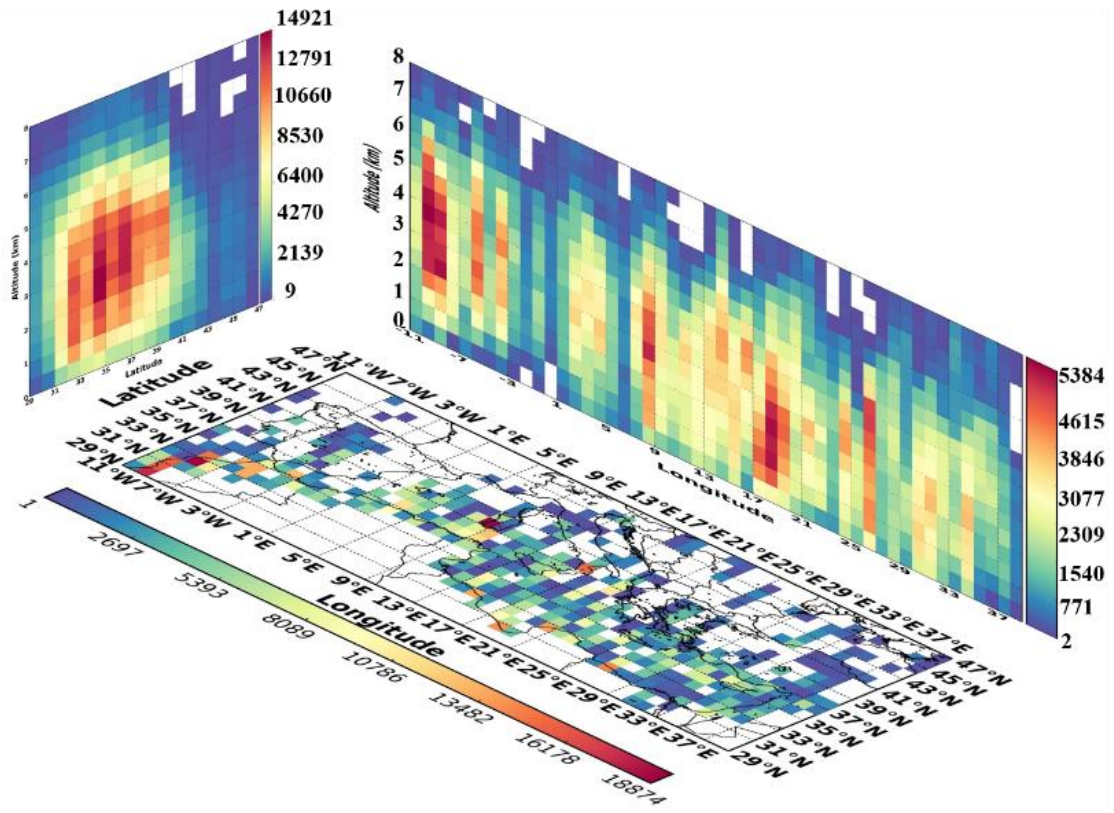
(ii-a)



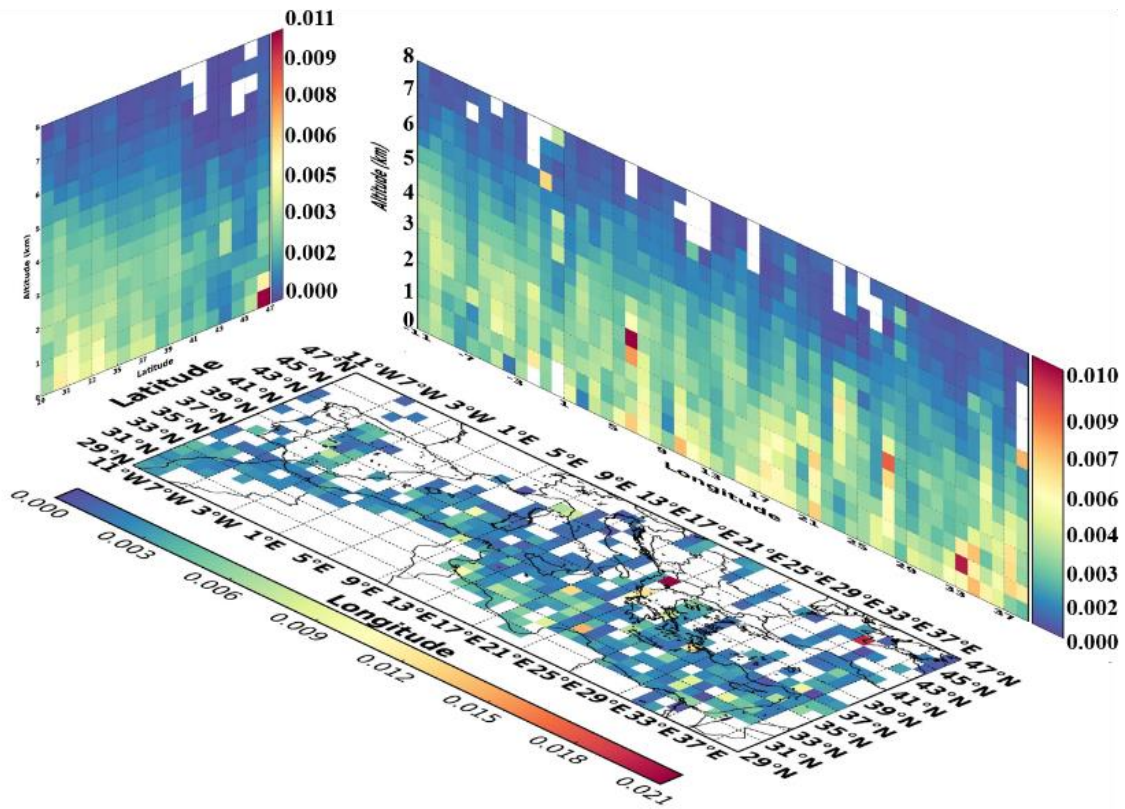
(ii-b)

**Figure 7:** Geographical distributions of the intensity (in terms of  $AOD_{550nm}$ ) of: (i) strong and (ii) extreme desert dust episodes, averaged over the periods: (a) Mar. 2000 – Feb. 2013 (MODIS-Terra) and (b) 2003 – 2012 (MODIS-Aqua), over the broader area of the Mediterranean basin.



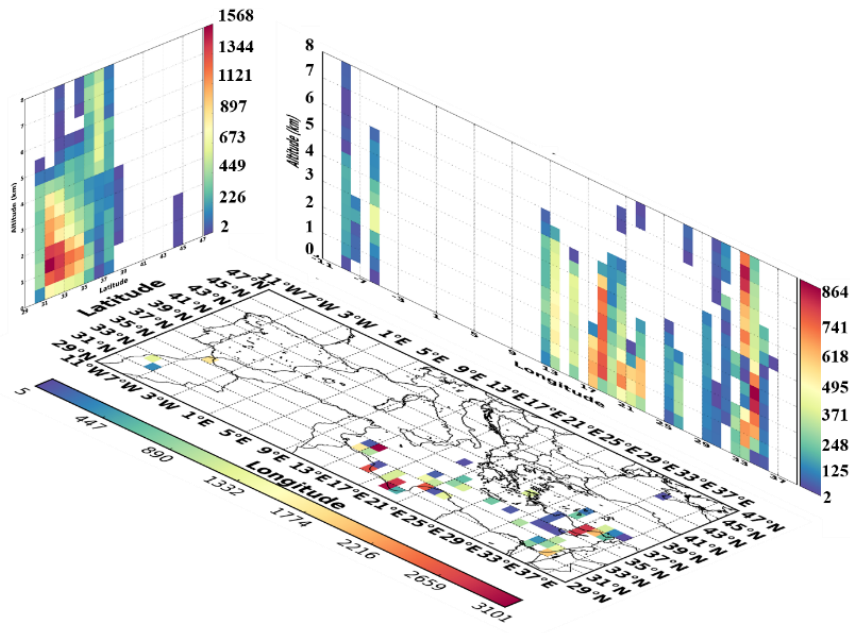


(i)

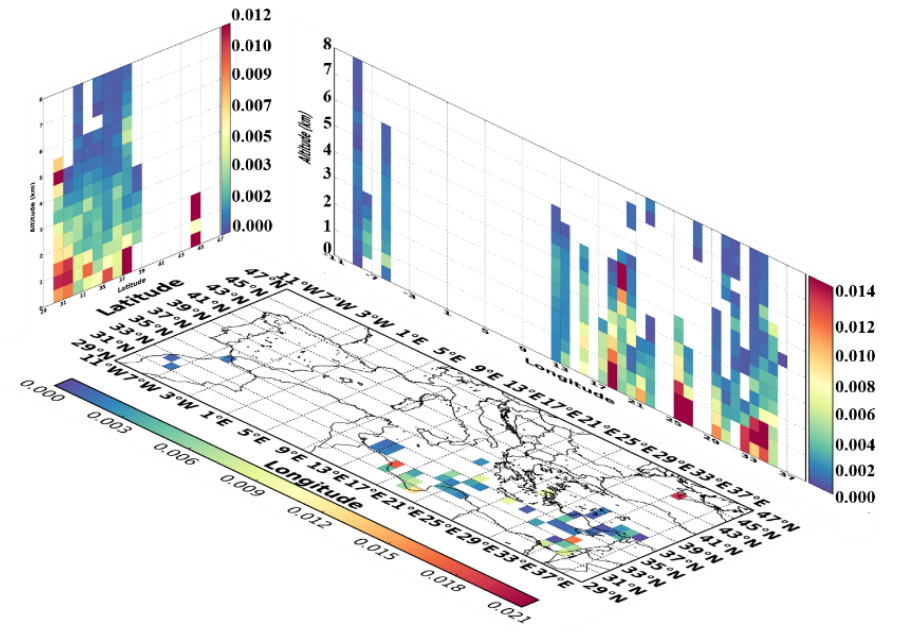


(ii)

**Figure 8:** Three dimensional structure of the: (i) overall number of dust and polluted dust observations and (ii) total backscatter coefficient at 532 nm (in  $\text{km}^{-1} \text{sr}^{-1}$ ), over the broader Mediterranean basin under DD episodes conditions, based on CALIOP-CALIPSO vertically resolved retrievals for the period Jun. 2006 – Feb. 2013.

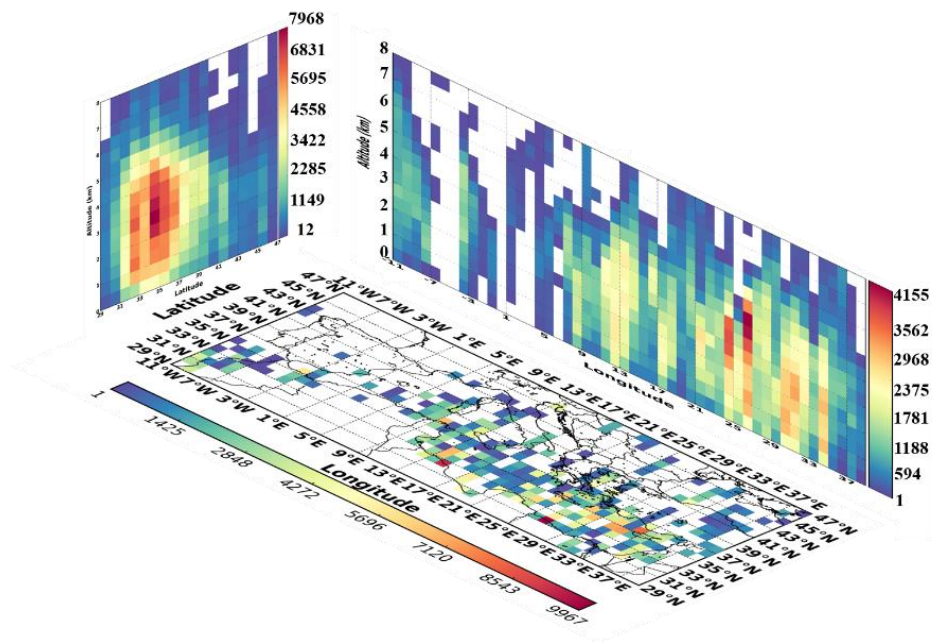


(i-a)

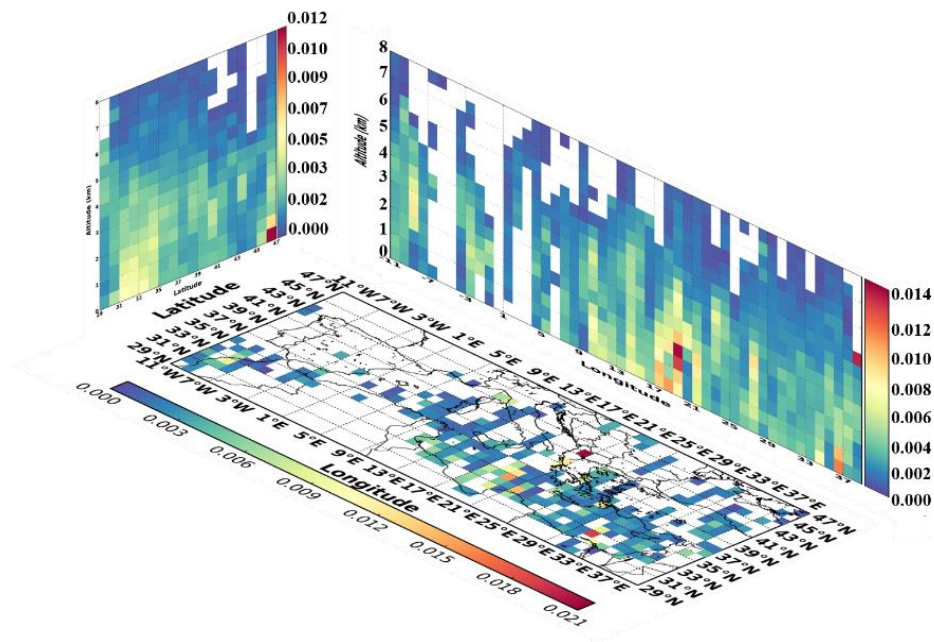


(i-b)

**Figure 9:** Three dimensional representation of the: (a) overall number of dust and polluted dust observations and (b) total backscatter coefficient at 532 nm (in  $\text{km}^{-1} \text{sr}^{-1}$ ), over the broader Mediterranean basin, under DD episodes conditions, for: (i) winter, (ii) spring, (iii) summer and (iv) autumn based on CALIOP-CALIPSO vertically resolved retrievals, over the period Jun. 2006 – Feb. 2013.



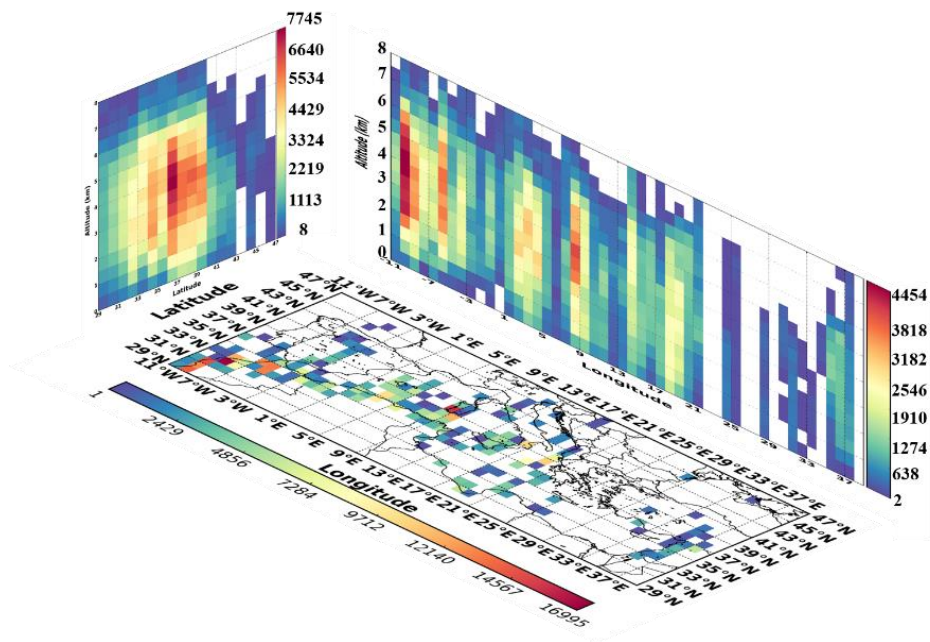
(ii-a)



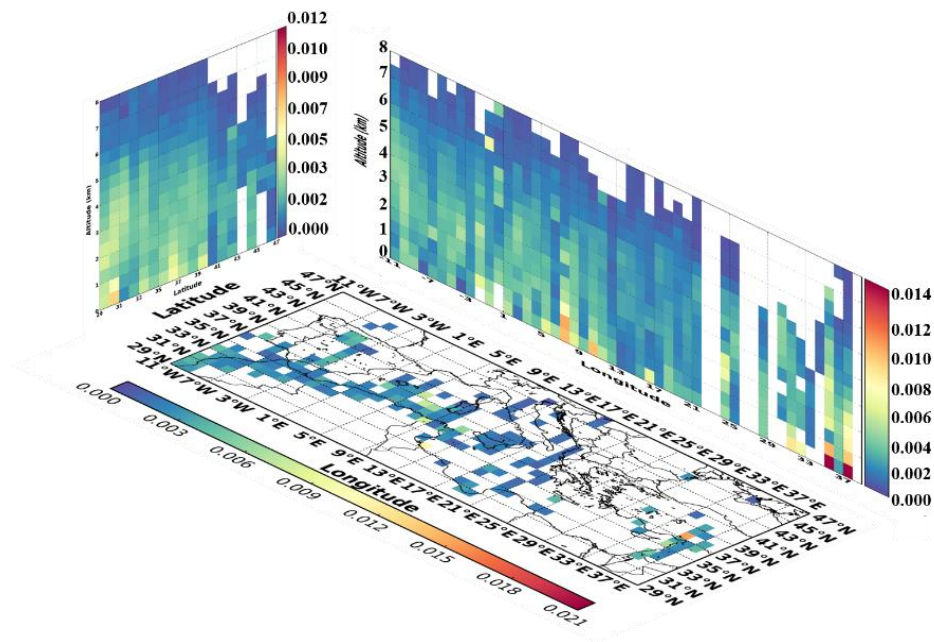
(ii-b)

**Figure 9:** Continued.



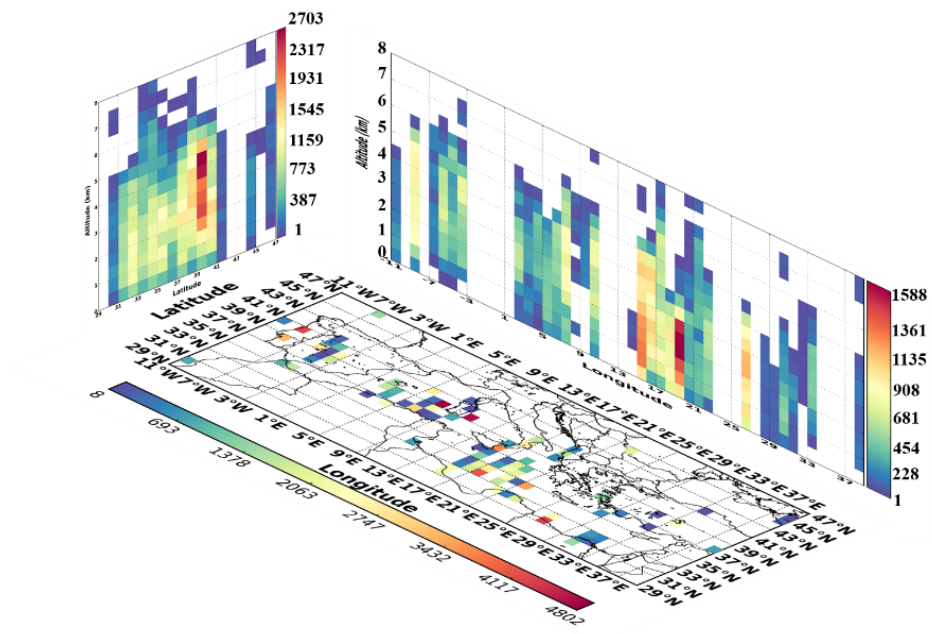


(iii-a)

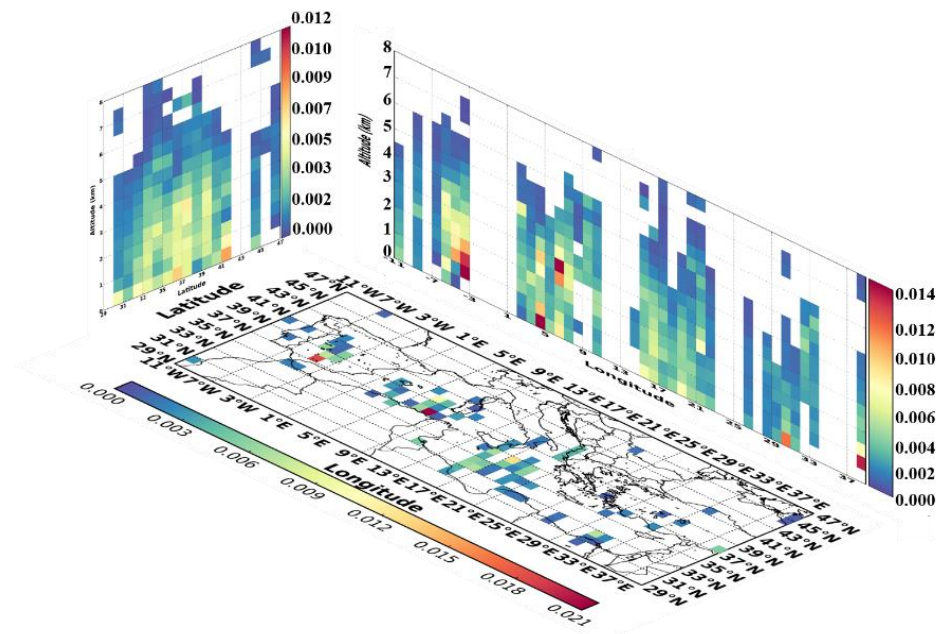


(iii-b)

Figure 9: Continued.



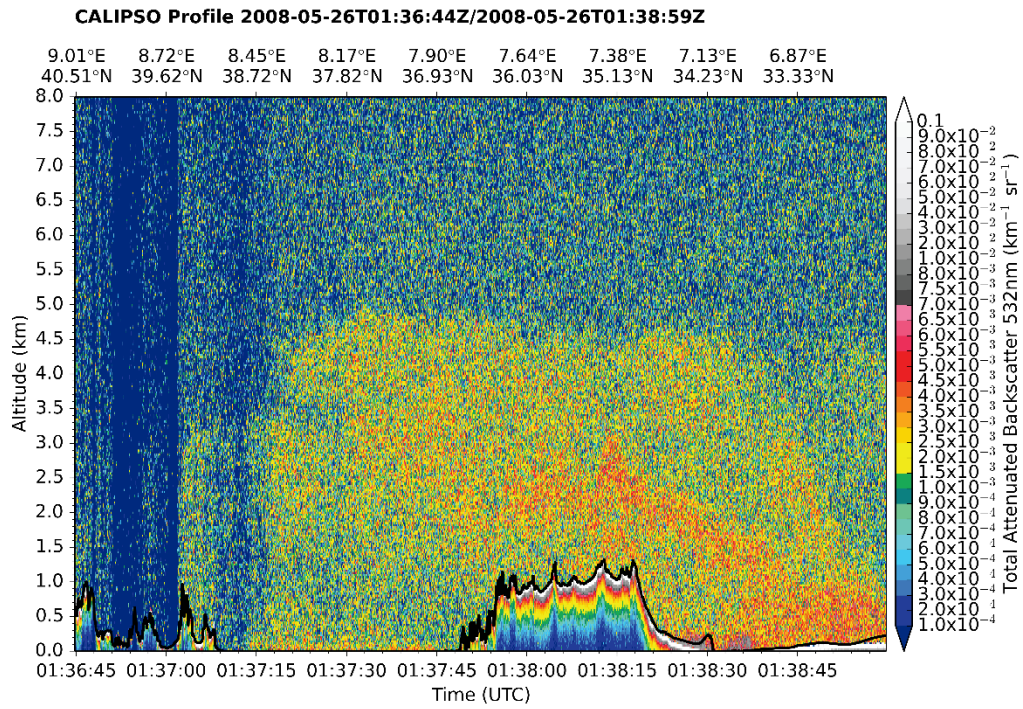
(iv-a)



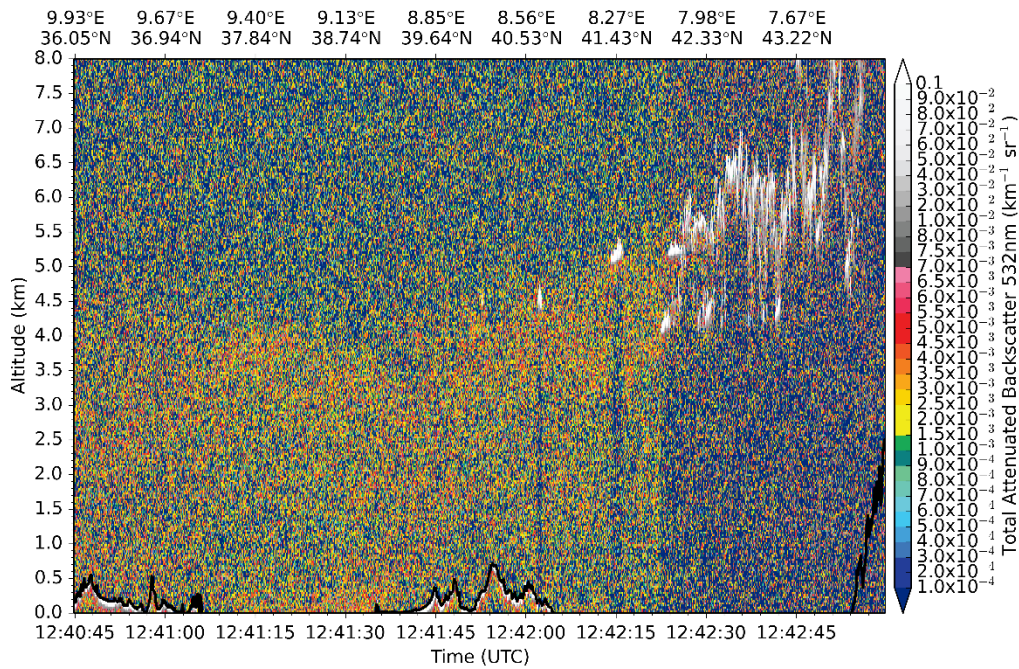
(iv-b)

**Figure 9:** Continued.



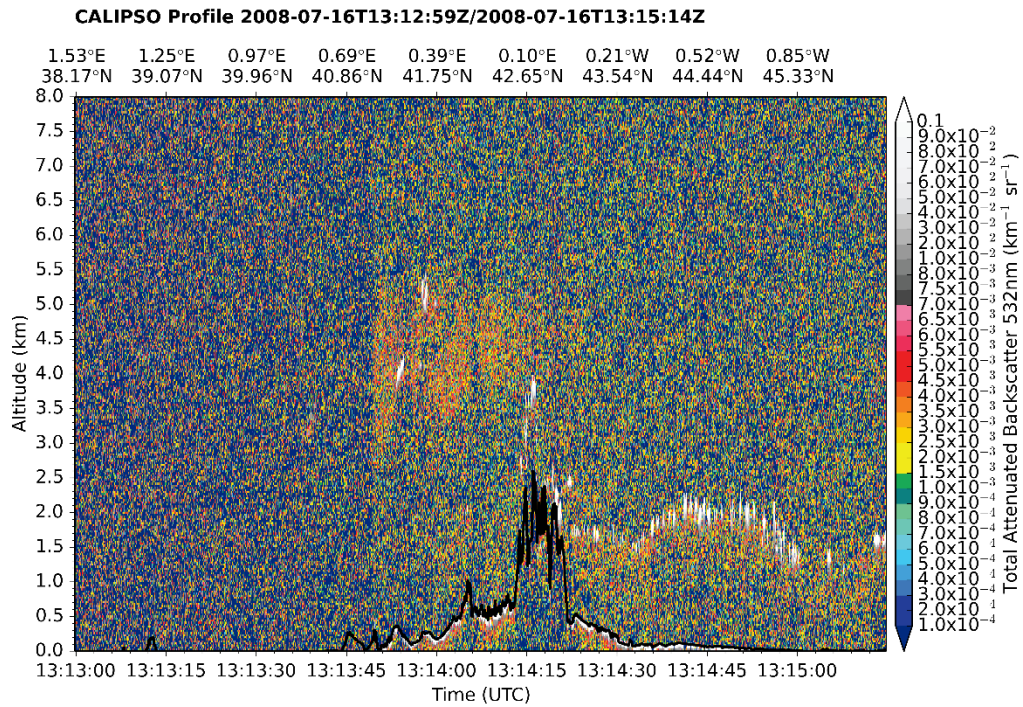


**CALIPSO Profile 2008-05-26T12:40:44Z/2008-05-26T12:42:59Z**

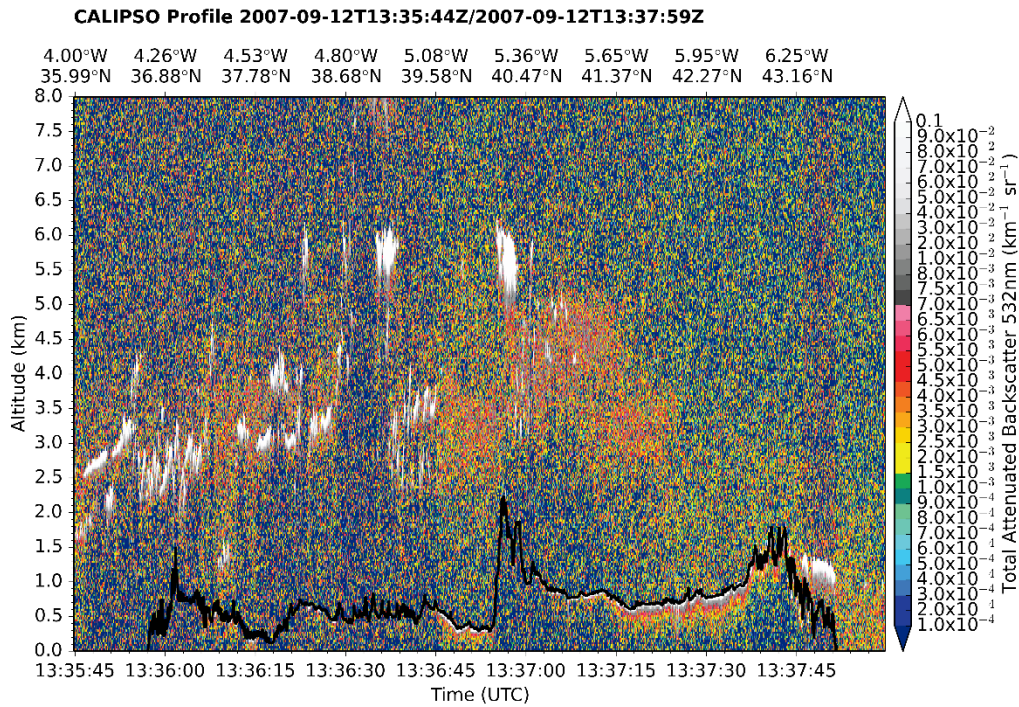


**Figure 10:** Cross sections of the total backscatter coefficient at 532 nm (in  $\text{km}^{-1} \text{sr}^{-1}$ ) vertical profiles along the CALIOP-CALIPSO track during: (i) nighttime and (ii) daytime, on 26<sup>th</sup> May 2008, over the station Censt (Lat: 39.064, Lon: 8.457). The black thick solid line represents the surface elevation.



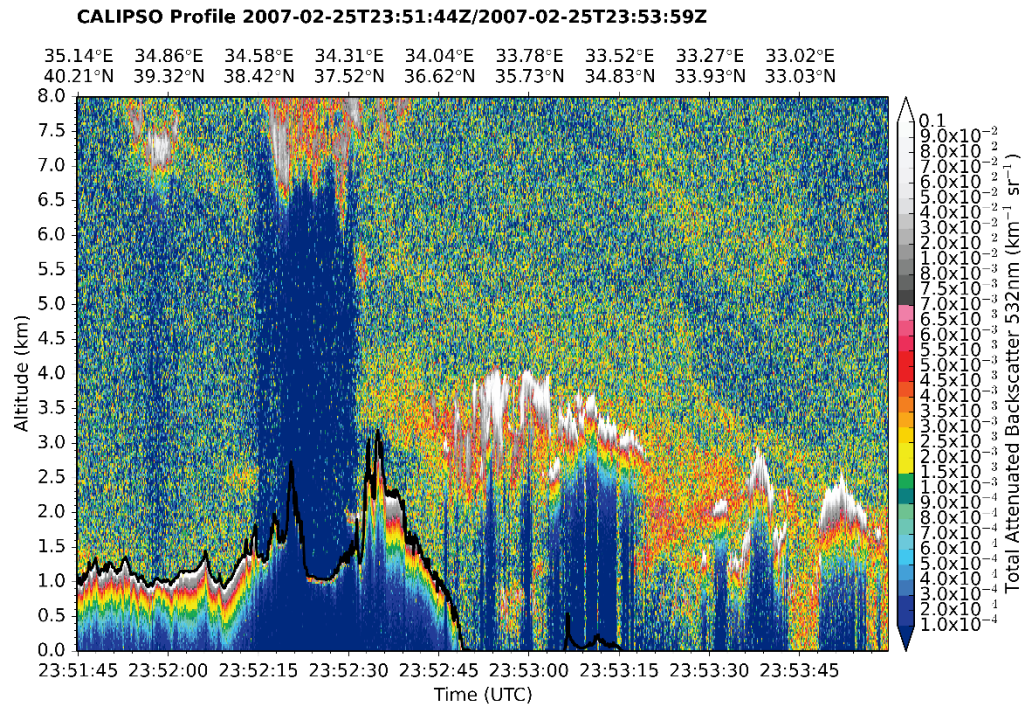


(i)



(ii)

**Figure 11:** Cross sections of the total backscatter coefficient at 532 nm (in  $\text{km}^{-1} \text{sr}^{-1}$ ) vertical profiles along the CALIOP-CALIPSO track during daytime over the stations: (i) Els Torms (Lat: 41.395, Lon: 0.721) on 16<sup>th</sup> July 2008 and (ii) San Pablo (Lat: 39.525, Lon: -4.353) on 12<sup>th</sup> September 2007. The black thick solid line represents the surface elevation.



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1852 **Figure 12:** Cross section of the total backscatter coefficient at 532 nm (in  $\text{km}^{-1} \text{sr}^{-1}$ ) vertical profiles along the CALIOP-  
 1853 CALIPSO track during nighttime over the station Agia Marina (Lat: 35.039, Lon: 33.058) on 25<sup>th</sup> February 2007. The black  
 1854 thick solid line represents the surface elevation.

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