

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) value 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 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 consistence 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. The CALIOP vertical profiles of pure and polluted dust observations and the associated total backscatter coefficient at 532 nm (β_{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, while 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 β_{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 β_{532nm} confirm the multilayered structure of the Mediterranean desert dust outbreaks on both annual and seasonal basis, 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 consideration of the dust vertical structure is necessary when attempting comparisons between columnar MODIS AOD retrievals and ground PM_{10} concentrations.

1. Introduction

The Mediterranean basin, due to its proximity to the major dust source arid areas of Northern Africa and Middle East (Middleton and Goudie, 2001; Prospero et al., 2002; Ginoux et al., 2012) is frequently affected by transported high dust loads referred to as episodes or events. The suspension and

accumulation of mineral particles into the atmosphere over the Saharan and Arabian Peninsula's deserts are determined by various factors such as the enhanced turbulence, soil conditions (reduced vegetation cover and soil moisture), reduced precipitation amounts, latitudinal shift of the Intertropical Convergence Zone (ITCZ) as well as by small scale meteorological processes (e.g. haboobs). However, dust particles can be transported far away from their sources, mainly towards the Atlantic Ocean (e.g. Prospero and Lamb, 2003; Ben-Ami et al., 2010; Huang et al., 2010) and Europe (e.g. Mona et al., 2006; Mona et al., 2012; Papayannis et al., 2008; Basart et al., 2012; Bègue et al., 2012; Pey et al., 2013), favored by the prevailing atmospheric circulation patterns, from planetary to synoptic scales. Due to their frequent transport in the Mediterranean, mineral dust particles, constitute the predominant aerosol type there (Barnaba and Gobbi, 2004; Basart et al., 2012), as shown by the good agreement, in spatial terms, between the geographical distributions of dust episodes' *AOD* (Gkikas et al., 2013) and average *AOD* conditions (Papadimas et al., 2008).

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

Extensive research has been also carried out on the mechanisms of Mediterranean dust outbreaks. Therefore, several mechanisms and processes of transport, apart from dust emissions in source areas,

98 have been proposed as controlling factors. Moulin et al. (1997) showed that the exported dust loads
99 from Northern Africa towards the Atlantic Ocean and the Mediterranean are controlled by the phase of
100 the North Atlantic Oscillation (NAO). Other studies, focused on the description of atmospheric
101 circulation characteristics favoring the occurrence of desert dust outbreaks over the central (Barkan et
102 al., 2005; Meloni et al., 2008) or western (Querol et al., 1998; Rodriguez et al., 2001; Salvador et al.,
103 2014) Mediterranean, but on a synoptic scale. An objective classification, based on multivariate
104 statistical methods, of the atmospheric circulation patterns related to dust intrusions over the
105 Mediterranean, has been presented by Gkikas et al. (2015) and Varga et al. (2014).

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

112 Dust transport over the Mediterranean is characterized by a multi-layered structure (Hamonou et
113 al., 1999; Papayannis et al., 2008) in contrast to the Atlantic Ocean, which is well confined to the
114 Saharan Air Layer (SAL, Karyampudi et al., 1999). The vertical distribution of dust load into the
115 troposphere as well as the profile of dust aerosols' optical properties at different altitudes, control the
116 impacts on atmospheric dynamics induced by the mineral particles (Zhang et al., 2013). In order to
117 describe the geometrical features of dust transport, many researchers have used ground lidar
118 measurements, model simulations (Alpert et al., 2004; Kishcha et al. 2005) or they have relied on a
119 synergistic use of satellite observations and ground lidar profiles (Berthier et al., 2006). The vertical
120 extension of the Saharan dust intrusions over Europe, during the period 2000-2002, was the subject of a
121 comprehensive study by Papayannis et al. (2008), who used lidar measurements from the EARLINET
122 (European Aerosol Research Lidar Network, Bösenberg et al., 2003). Over the Mediterranean stations,
123 the mean base, top and thickness of dust layers was found to vary from 1356 to 2980 m, 3600 to 5900
124 m and 726 to 3340 m, respectively. According to the obtained results, tracers of dust particles can be
125 detected up to 10 km, as also reported by Gobbi et al. (2000), who studied a Saharan dust event in
126 Crete (south Greece) during spring of 1999.

127 Several similar studies have been also performed for specific Mediterranean locations based on
128 EARLINET lidar measurements. For example, Mona et al. (2006) analyzed the vertical structure of 112

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

146 Surface-based lidar measurements like those used in the aforementioned studies provide useful
147 information about the geometrical and optical properties of dust layers, but they are representative only
148 for specific locations. Yet, a more complete knowledge about the vertical structure of dust outbreaks is
149 necessary in order to adequately understand and determine their possible effects. The limitation
150 imposed by the use of surface-based lidar observations can be overcome by utilizing accurate satellite
151 retrievals, as a complementary tool, which provide extended spatial coverage. Since 2006, vertical
152 resolved observations of aerosols and clouds from space were made possible thanks to the CALIOP
153 (Cloud-Aerosol Lidar with Orthogonal Polarization) lidar flying onboard the CALIPSO (Cloud-
154 Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite (Winker et al., 2009). Based on
155 CALIOP observations, Liu et al. (2008) analyzed the global vertical distribution of aerosols for one
156 year, while other studies focused on the vertical structure of dust outflows towards the Atlantic Ocean
157 (e.g. Ben-Ami et al., 2009; Adams et al., 2012; Tsamalis et al., 2013) and the Pacific Ocean (e.g.
158 Eguchi et al., 2009; Hara et al., 2009). On the contrary, over the broader Mediterranean area, only a
159 small number of studies has been made aiming at describing the vertical distribution of dust aerosols
160 (Amiridis et al., 2013) or specifying the vertical structure of dust events (Amiridis et al., 2009).

161 Nevertheless, they only dealt with a single dust event (18-23 May 2008, Amiridis et al., 2009) and thus
162 cannot satisfy the need to know the general vertical structure of Mediterranean dust episodes.

163 The main target of the present study is to describe the Mediterranean desert dust outbreaks' vertical
164 structure. For this purpose, satellite retrievals derived by the MODIS-Terra/Aqua, Earth Probe-TOMS,
165 OMI-Aura and CALIOP-CALIPSO databases (Section 2) are used in a synergistic way. The dust
166 outbreaks are identified with an objective and dynamic algorithm, which uses appropriate aerosol
167 optical properties representative of suspended particles' load, size and nature (Section 3). Based on its
168 outputs, the primary characteristics of the intense Mediterranean desert dust (DD) episodes, namely
169 their frequency and intensity, are described in Section 4.1. Just in order to assess the consistency of the
170 algorithm' concept, an alternative methodology for the determination of DD episodes is also applied
171 and the obtained results are inter-compared with the basic methodology. The outputs of the default
172 version of the satellite algorithm are compared versus surface measurements provided by AERONET
173 or PM₁₀ stations, located within the study region (Section 4.2). Additionally, useful information about
174 various optical and physical properties under intense dust episodes conditions is also derived from the
175 aforementioned analysis. For the identified DD episodes, collocated CALIOP-CALIPSO vertical
176 feature mask and total backscatter coefficient at 532 nm retrievals are used in order to describe the
177 annual and seasonal variability of dust outbreaks' vertical extension over the Mediterranean (Section
178 4.3). Moreover, in Section 4.4, a thorough analysis of few specific Mediterranean DD episodes is
179 made, in order to examine how the vertical distribution of desert dust outbreaks can affect the
180 agreement between MODIS AOD and PM₁₀ data. Finally, the summary and conclusions are drawn in
181 Section 5.

182

183 **2. Satellite and surface-based data**

184 The different types of satellite retrievals that have been used as inputs to the objective and dynamic
185 satellite algorithm are described below, namely the MODIS (Section 2.1.1), EP-TOMS and OMI-Aura
186 (Section 2.1.2) databases. Also, CALIOP-CALIPSO vertically resolved satellite data, coincident with
187 the identified desert dust outbreaks by the satellite algorithm, are described in Section 2.1.3. Finally,
188 surface-based sun-photometric AERONET retrievals and PM₁₀ concentrations, both used for the
189 comparison against the satellite algorithm's outputs, are described in Sections 2.2.1 and 2.2.2,
190 respectively.

191

192 2.1 Satellite data

193 2.1.1 MODIS

194

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

207 The following daily MODIS-Terra and MODIS-Aqua Collection 051 (C051) level 3 satellite data
208 (MOD08_D3 and MYD08_D3 files) provided at $1^\circ \times 1^\circ$ latitude-longitude spatial resolution are used:
209 (i) AOD_{550nm} , (ii) Ångström exponent over land ($\alpha_{470-660nm}$), (iii) Ångström exponent over ocean
210 ($\alpha_{550-865nm}$), (iv) fine-mode fraction (*FF*) of *AOD* over land and ocean and (v) Effective radius over
211 ocean (r_{eff}). It must be mentioned that the size parameters (α , *FF*) over land are less reliable compared
212 to the corresponding ones over sea, since they are highly sensitive to spectral dependent factors such as
213 errors in the surface model or sensor calibration changes. Over sea, the accuracy of size parameters is
214 strongly dependent on wind conditions. Similar data have been used by Gkikas et al. (2013), however,
215 in the present study we have improved data quality by using the quality assurance-weighted (QA) level
216 3 data (http://modis-atmos.gsfc.nasa.gov/docs/QA_Plan_2007_04_12.pdf) derived from the level 2
217 retrievals (10 km x 10 km spatial resolution). Each level 2 retrieval, is flagged with a bit value (from 0
218 to 3) corresponding to confidence levels (No confidence: 0, Marginal: 1, Good: 2 and Very Good: 3).
219 Based on this, the level 3 QA-weighted spatial means are obtained by the corresponding level 2
220 retrievals considering as weight their confidence level (bit value). In addition, the day cloud fraction as
221 well as the number of level 2 counts, which are both relevant to the performance of the satellite

algorithm, are also used in this study. The time series of daily MODIS aerosol data cover the 13-yr period March 2000-February 2013 (Terra) and the 10-yr period January 2003-December 2012 (Aqua).

2.1.2 EP/TOMS and OMI-Aura

The selected retrievals from MODIS provide information about particles' load (*AOD*) and size (α , FF , r_{eff}), which are both necessary to identify dust episodes. However, since dust is not the only coarse aerosol, for example sea-salt can be so as well, another optical property indicative of particle absorption efficiency is also required by the algorithm. To address this issue, the Absorption Aerosol Index (*AI*) daily data were also used, derived from measurements taken by the Total Ozone Mapping Spectrometer (TOMS) instrument onboard the NASA's Earth-Probe satellite (2000-2004) and the Ozone Monitoring Instrument (OMI) onboard the NASA's Aura satellite (2005-2013). *AI* is the primary TOMS aerosol product (Herman et al., 1997) based on a spectral contrast method in a UV region (331-360 nm) where ozone absorption is very small and can be used for the distinction between scattering (e.g. sea-salt) and absorbing (e.g. desert dust, smoke) aerosols. The retrieval algorithm (fully described by Torres et al., 1998; 2002; 2005) takes advantage of the low surface albedo in the UV spectrum range, even in arid and semi-arid areas, making thus possible the estimation of the *AOD* over highly reflecting desert surfaces, where the major dust sources are located. Since the late 70's, the TOMS sensor onboard Nimbus-7 (1978 – 1993) and Earth Probe (1996 – 2005) has been providing global aerosol measurements. With the deployment of the EOS-Aura OMI (Ozone Monitoring Instrument) in mid-2004 (Torres et al., 2007) the near UV aerosol record continues to be extended into the foreseeable future. OMI is a hyperspectral sensor, covering the 270-500 nm range, launched onboard the EOS-Aura satellite on July 15, 2004 (1:38 pm equator crossing time, ascending mode) providing almost daily global coverage thanks to its wide viewing swath (2600 km with 13 km x 24 km nadir resolution). Apart from *AI* measurements, OMI aerosol products include also the total and absorption *AOD* and the single scattering albedo at 388 and 500 nm (Torres et al., 2007). Both EP-TOMS and OMI-Aura retrievals are available via the Mirador ftp server (<http://mirador.gsfc.nasa.gov/>) of the Goddard Earth Sciences Data and Information Services Center (GES DISC). OMI-Aura data, as MODIS, are provided at 1° x 1° spatial resolution while the EP-TOMS retrievals have been regridded from their raw spatial resolution (1° x 1.25°) in order to match with the other two datasets (OMI, MODIS).

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252 2.1.3 CALIOP-CALIPSO

253

254 The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard the NASA's satellite
255 CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations), launched in April
256 2006, provides vertical resolved aerosol and cloud observations (Winker et al., 2009) since June 2006.
257 CALIPSO is flying in the A-Train constellation (Stephens et al., 2002; <http://atrain.nasa.gov/>) in a sun-
258 synchronous polar orbit at 705 km over the surface, with a 16-day repeat cycle, crossing the equatorial
259 plane at about 13:30 local solar time (Winker et al., 2009). CALIOP is an active sensor measuring the
260 backscatter signal at 532 nm and 1064 nm as well as the polarization at 532 nm (Winker et al., 2009).
261 These level 1 retrievals are further processed (calibration and range corrections) passing to Level 2 in
262 order to retrieve the backscatter and extinction coefficients, at 532 nm and 1064 nm, for aerosol and
263 cloud layers. The identification of cloud and aerosol layers within the atmosphere (Vaughan et al.,
264 2009) is made through the cloud aerosol discrimination (CAD) algorithm (Liu et al., 2009), which is
265 based on the probability distribution functions (PDFs) of altitude-and-latitude-dependent parameters
266 (integrated color ratio, layer-integrated volume depolarization ratio, mean attenuated backscatter
267 coefficient). CAD scores vary mainly from -100 to 100 indicating the presence of aerosols and clouds
268 when are negative and positive, respectively, while bins of confidence levels, both for aerosols and
269 clouds, are defined based on their absolute values
270 ([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)
271 [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 VFM
272 algorithm, either for aerosols or clouds, is more reliable for increasing CAD scores in absolute terms.
273 Aerosols are categorized in 6 primary types namely: (i) clean marine, (ii) dust, (iii) polluted
274 continental, (iv) clean continental, (v) polluted dust and (vi) smoke (Omar et al., 2009).

275 In the present analysis, we use the Version 3 (3.01 and 3.02) of the Level 2 Vertical Feature Mask
276 (VFM) and Aerosol Profile Products (APro) files, available from June 2006 to February 2013, both
277 derived from the NASA's Earth Observing System Data and Information System
278 (<http://reverb.echo.nasa.gov/>). The aerosol profile products are generated at a uniform horizontal
279 resolution of 5 km (http://www-calipso.larc.nasa.gov/products/CALIPSO_DPC_Rev3x6.pdf), while the
280 vertical resolution varies from 60 to 180 m depending on the altitude range and the parameter. The
281 scientific data sets which have been analyzed are the following: (i) aerosol subtype, (ii) CAD score and

282 (iii) Total Backscatter Coefficient at 532 nm (β_{532nm}), reported at several tropospheric and stratospheric
283 levels above mean sea level (Hunt et al., 2009).

284

285 2.2 Surface-based data

286

287 2.2.1 AERONET

288 The AErosol RObotic NETwork (AERONET, Holben et al., 1998) is a worldwide network of
289 installed CIMEL sun-sky radiometers obtaining sun-photometric observations in more than 1000
290 locations of the planet (<http://aeronet.gsfc.nasa.gov>). The solar irradiances received by the photometer
291 are inversed to columnar aerosol optical and microphysical properties through the implementation of
292 retrieval algorithms (e.g. Dubovik and King, 2000; O' Neill et al., 2003). The followed standardized
293 methods concerning instrument maintenance, calibration, cloud screening and data processing allow
294 aerosol monitoring and comparison between different study periods and areas (Smirnov et al., 2000).
295 From the global AERONET stations, 109 are located within the geographical limits of our study
296 region. For each station, the daily averages of cloud-screened and quality assured data (Level 2.0) of
297 direct sun and almucantar retrievals are used for: (i) *AOD* at 7 wavelengths from 340 to 1020 nm, (ii)
298 size distribution retrieved for 22 logarithmically equidistant discrete points (r_i) in the range of sizes
299 $0.05 \mu\text{m} \leq r \leq 15 \mu\text{m}$, (iii) Ångström exponent between 440 and 870 nm ($\alpha_{440-870nm}$), (iv) total effective
300 radius (r_{eff}), and (v) single scattering albedo (SSA) and asymmetry parameter (g_{aer}) both retrieved at 440
301 nm, 675 nm, 870 nm and 1020 nm. The uncertainty in the estimation of *AOD* depends on technical
302 (e.g. calibration method) factors and inversion assumptions, both described in detail in Holben et al.
303 (1998). Moreover, the accuracy of the retrieved *AOD* by the CIMEL radiometer is spectrally
304 dependent, being higher ($<\pm 0.01$) for wavelengths longer than 440 nm and lower ($<\pm 0.02$) for the UV
305 wavelengths (Eck et al., 1999). It should be also noted that the AERONET Level 2.0 inversion products
306 (e.g. SSA) are provided when *AOD* at 440 nm is higher than 0.4 ensuring the minimization of the
307 inversion uncertainties, which are also determined by other factors (e.g. scattering angle, particles'
308 sphericity) as stated in detail by Dubovik et al. (2000).

309

310 2.2.2 PM_{10}

311 Daily total and dust surface PM_{10} concentrations, over the period 2001-2011 from 22 regional
312 background and suburban background sites were used in this study. The monitoring sites are distributed

as follows: 10 in Spain; 2 in southern France; 5 in Italy; 3 in Greece; 1 in southern Bulgaria and 1 in Cyprus. PM₁₀ concentrations were obtained in most cases from gravimetric determinations on filters, whereas in few cases they were determined by real time instruments (Querol et al., 2009b; Pey et al., 2013) but corrected against gravimetric measurements carried out in annual field campaigns. The disaggregation of the dust component to the total amount is made based on a statistical approach which has been applied in several past studies (e.g. Rodríguez et al., 2001; Escudero et al., 2007; Querol et al., 2009b; Pey et al., 2013). A full description of the methodology which is followed for the calculation of dust particles' contribution to the total PM₁₀ is presented in Escudero et al. (2007). Briefly, the net dust PM₁₀ amount is calculated through the subtraction of the regional background PM₁₀, which is obtained by applying a monthly moving 30th percentile to the PM₁₀ timeseries excluding days of dust transport, from the corresponding values of the total PM₁₀ concentrations. Most of the derived data were obtained from the AirBase (<http://acm.eionet.europa.eu/databases/airbase/>) database, while for the stations Finokalia (Crete) and Montseny (NE Spain) the relevant measurements have been acquired from the EUSAAR (<http://www.eusaar.net/>) database.

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328 **3. Identification of desert dust episodes**

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Following the methodology proposed by Gkikas et al. (2013), desert dust (DD) episodes are identified based on an objective and dynamic algorithm, which is depicted in the flowchart of Figure 1. The algorithm operates in three steps and is applied in each individual 1° x 1° geographical cell within the geographical limits of the study domain (29° N - 47° N and 11° W - 39° E). First (Fig. 1, yellow box), the mean (*Mean*) and the associated standard deviation (*Std*) from the available *AOD*_{550nm} retrievals are calculated for the whole study period. These primary statistics are used for the definition of two threshold levels, which are equal to *Mean*+2**Std* and *Mean*+4**Std*. 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* (cyan boxes). 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 *AOD*s, also possibly relevant to radiative and health effects, are masked out from the

345 dataset. In order to investigate the possible impact of this, “unbiased” mean, standard deviation and
346 thresholds of AOD are also computed based on another methodology and the results are discussed
347 comparatively to those of the primary methodology in a separate paragraph. Moreover, it must be
348 mentioned that the satellite algorithm identifies only intense desert dust episodes since their *AOD* must
349 be higher than $Mean+2*Std$ which is considered as a high threshold level.

350 It should be noted that the representativeness of the calculated mean levels is possibly affected by
351 the availability of the AOD retrievals and particularly by the way these data are distributed at different
352 temporal scales. To this aim, we have calculated the percentage availability of AOD retrievals on a
353 monthly, seasonal and year by year basis, over the period 2000-2013 (results not shown here). Seasonal
354 differences of AOD availability are mainly encountered in the northern parts of the study region, with
355 lower values (20 to 40 %) from December to February against 50-85% for the rest of the year. This is
356 attributed to the enhanced cloud coverage prohibiting the satellite observations. Nevertheless, this does
357 not essentially affect the algorithm outputs since these regions, being far away from the dust sources,
358 are not so frequently affected by dust outbreaks, especially given the significant wet removal of
359 aerosols during this most rainy season of the year. On a year by year basis, the differences of the AOD
360 data’s availability are almost negligible.

361 In a further step of the methodology, the strong and extreme DD episodes are identified separately
362 over land and sea surfaces of the study region. This is achieved through the usage of specific aerosol
363 optical properties, namely the Ångström exponent, effective radius, fine fraction and aerosol index,
364 which provide information about particles’ size and nature (black box, Figure 1). For each optical
365 property, appropriate upper or lower thresholds have been set up (green boxes, Figure 1) which must be
366 valid concurrently in order to certify the presence of dust particles in the atmosphere. These cut-off
367 levels have been selected according to the literature findings, availability of raw data and several
368 sensitivity tests (more details are provided in Gkikas et al., 2013). The validity of these thresholds is
369 further evaluated against AERONET measurements and the corresponding results are discussed in
370 Section 4.2.1.4.

371 In order to address the issue of possible overestimation of the defined threshold levels, particularly
372 in the most dust affected areas as it has been mentioned above, we have also applied the satellite
373 algorithm using an alternative methodology (METHOD-B) in which dust-affected grid cells were
374 excluded. In this case, from the raw AOD retrievals we have masked out the “pure” desert dust grid
375 cells, which were identified based on the concurrent fulfillment of the defined criteria for dust
376 occurrence in the algorithm (for Ångström exponent, fine fraction, aerosol index and effective radius,

377 green boxes of Figure 1). Then, from the remaining data (non-dust AOD retrievals), the mean, the
378 associated standard deviation as well as the defined thresholds of AOD are computed for the whole
379 study period, for each pixel, as also done in the primary methodology. Finally, also similarly to the way
380 done in the primary methodology, the DD episodes were classified into strong and extreme ones. The
381 frequency of occurrence and intensity of DD episodes determined with METHOD-B are provided in
382 the supplementary material (Figures S1 and S2) while their differences with regards to the primary
383 methodology are discussed in Section 4.1.

384 As explained, a similar methodology and data were used in the study by Gkikas et al. (2013).
385 Nevertheless, the present one is a significant extension mainly for five reasons: (i) DD episodes are
386 identified here over an extended period of study and for both MODIS platforms, i.e. Mar. 2000 – Feb.
387 2013 for MODIS-Terra and 2003-2012 for MODIS-Aqua, (ii) a second methodology (METHOD-B)
388 for the identification of DD episodes is tested, (iii) the quality of the input data is improved by using
389 QA-weighted level-3 data produced by weighting level-2 data based on their confidence flag instead of
390 regular ones ($QA \geq 1$), (iv) emphasis is given to the vertical structure of the intense DD episodes and (v)
391 the role of the detailed dust outbreaks' vertical structure for the level of agreement between columnar
392 MODIS AOD and ground PM_{10} concentrations is investigated. Moreover, an improvement of the
393 methodology consists in the application of our satellite algorithm also using only AODs associated with
394 cloud fractions (CF) lower/equal than 0.8, in order to investigate possible modifications of our results
395 due to the cloud contamination effects on MODIS AODs. The critical value of 0.8 for CF has been
396 defined according to Zhang et al. (2005) and Remer et al. (2008), who stated that under extended cloud
397 coverage conditions AOD levels can be increased substantially.

398

399

400 4. Results

401 Before dealing with the vertical structure of dust outbreaks (sub-sections 4.3 and 4.4), it is very
402 important to describe their horizontal patterns (sub-section 4.1) and also to compare the algorithm's
403 outputs against quality AERONET and PM_{10} observations (sub-section 4.2) in order to ensure an
404 accurate three-dimensional view of the intense Mediterranean DD episodes. It must be clarified, that
405 the comparison of the satellite algorithm's outputs versus AERONET/ PM_{10} is made only for its default
406 version and not for the METHOD-B, since between the two methodologies are not found remarkable
407 differences, as it will be presented in Section 4.1. Accordingly, the synergistic implementation of the
408 CALIOP-CALIPSO lidar profiles is done only when the DD episodes are identified based on the

primary methodology. The present section has been organized accordingly and the results are given below.

411

4.1 2D geographical distributions of desert dust episodes' frequency and intensity

The mean geographical distributions of strong and extreme DD episodes' frequency of occurrence (episodes yr^{-1}) are presented in Figure 2. 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. Marriotti et al., 2002; Mehta and Yang, 2008).

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

440 discrepancy appeared between MODIS-Terra and MODIS-Aqua spatial distributions, is attributed to
441 the diurnal variation of factors regulating the emission and transport of dust particles from the sources
442 areas. Schepanski et al. (2009), analyzed the variation of the Saharan dust source activation throughout
443 the day, based on MSG-SEVIRI satellite retrievals, reporting that dust mobilization is more intense in
444 the local early morning hours after sunrise. Note, that desert dust episodes over the period Mar. 2000 –
445 Feb. 2013 have been identified based on observations retrieved by the Terra satellite, which flies over
446 the study region around noon in contrast to Aqua which provides aerosol measurements at early
447 afternoon hours.

448 The analysis has been also repeated (results not shown here) considering as inputs to the satellite
449 algorithm only *AODs* associated with cloud fractions lower/equal than 0.8, in order to investigate
450 possible modifications to our results (Figs 2 and 3) due to the cloud contamination effect. As it
451 concerns the strong DD episodes, the geographical distributions are similar with those of Fig. 2, but the
452 maximum frequencies (recorded in Morocco) are higher by up to 2 episodes yr^{-1} and 0.3 episodes yr^{-1}
453 for the MODIS-Terra (Mar. 2000 – Feb. 2013) and MODIS-Aqua (2003-2012) data set, respectively.
454 On the contrary, in the case of extreme DD episodes the maximum frequencies decrease to 2.5 episodes
455 yr^{-1} for the period 2003-2012 and they shift southwards, namely over the northern coasts of Africa,
456 while over the central parts of the Mediterranean Sea are lower than 1 episode yr^{-1} .

457 The maps of intensities (in terms of AOD_{550nm}) of DD episodes (Figure 3), show that for both study
458 periods and satellite platforms, the maximum intensities are over the Gulf of Sidra and the Libyan Sea,
459 along the northern African coasts. These intensities reach *AODs* up to about 1.5 for strong and 4.1 for
460 extreme episodes, while the minimum ones (values down to 0.25-0.46) are recorded in the northern and
461 western Mediterranean parts. Note that dissimilar spatial patterns appear between the geographical
462 distributions of DD episodes' frequency and intensity, indicating that these two features are determined
463 by different factors (e.g. tracks or strength of depressions). Finally, when the cloud contamination is
464 minimized using only *AODs* associated with *CF* lower than 0.8, then the maximum intensities are
465 shifted southwards, across the northern Africa and eastern coasts of the Mediterranean, being lower
466 than 1 and 2 for strong and extreme DD episodes, respectively. Through the rejection of possibly
467 overestimated *AODs* from the dataset, it is found that the threshold levels are decreased (mainly over
468 the most frequently dust affected areas) since both mean and standard deviation values are lower
469 (results not shown here). Nevertheless, even though these *AODs* can be overestimated, in the majority
470 of the cases the collocated AERONET *AODs* are high (but lower than the satellite observations)
471 indicating the occurrence of desert dust outbreaks as it will be shown in Section 4.2.1.4.

472 The analysis has been also repeated applying the alternative METHOD-B described in Section 3.
473 Just to ensure a longer temporal coverage, this analysis was done for the period Mar. 2000-Feb. 2013
474 using MODIS-Terra data. The obtained results for the frequency of occurrence as well as for the
475 intensity of DD episodes are depicted in Figures S1 and S2, respectively, in the supplementary
476 material. The geographical patterns for the frequency of occurrence between the two methodologies are
477 similar; however, the maximum values for the strong and extreme DD episodes can reach up to 13.3
478 episodes year⁻¹ (Fig. S1-i) and 8.1 episodes year⁻¹ (Fig. S1-ii), respectively. As it concerns the intensity,
479 the geographical patterns, particularly for the strong DD episodes, are dissimilar and less distinct
480 compared to the corresponding ones obtained with the primary methodology. This difference is
481 attributed to the inclusion of more dust episodes with variable intensity, which leads to a not so clear
482 “signal” when all these episodes are averaged. Based on METHOD-B, the maximum intensities (in
483 terms of AOD_{550nm}) of strong DD episodes can reach up to 1 (Fig. S2-i) while for the extreme episodes
484 (Fig. S2-ii) it can be as large as 3. The main finding, based on the intercomparison of the two
485 methodologies for the identification of DD episodes, is that the frequency of the episodes is higher for
486 the METHOD-B with respect to the primary methodology, while the intensity is decreased. Both facts
487 are expected and can be explained by the lower calculated AOD thresholds with METHOD-B thus
488 yielding more DD episodes of lower intensity.

489 This introductory analysis was conducted in order to specify the locations where the Mediterranean
490 dust outbreaks occur more frequently and are more intense. Nevertheless, this paper is orientated to the
491 description of the intense Mediterranean dust outbreaks’ vertical structure as well as to the detailed
492 assessment of the applied satellite algorithm for the identification of DD episodes in order to
493 consolidate our methodology, and not to emphasize on their regime, which has been thoroughly
494 analyzed in Gkikas et al. (2013).

495

496 4.2 Comparison of the satellite algorithm’s outputs against AERONET and PM₁₀ measurements

497 The ability of the satellite algorithm to identify satisfactorily DD episodes, is tested against ground
498 measurements from 109 AERONET (Fig. 4, orange squares) and 22 PM₁₀ (Fig. 4, green triangles)
499 stations located in the broader Mediterranean area. This is an extended and thorough comparison which
500 exceeds largely a similar one done for the outputs of the previous version of satellite algorithm (2000-
501 2007, Gkikas et al., 2013), but only relying on 9 AERONET stations and using AOD and volume size
502 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 4, 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 4) the intense DD episodes have been identified from ground (AERONET) and the corresponding results are compared with the satellite algorithm outputs (Section 4.2.1.4). Finally, the performance of the algorithm is also tested against surface PM_{10} measurements from 22 stations (Section 4.2.2).

4.2.1 AERONET

4.2.1.1 Aerosol optical depth

During the period Mar. 2000 – Feb. 2013, 346 pixel level intense DD episodes have been identified by the satellite-based algorithm, in which coincident MODIS-Terra and AERONET retrievals are available. It should be noted that AERONET AOD_{550nm} values have been calculated from available AERONET AOD_{870nm} and Ångström exponent data ($\alpha_{440-870nm}$) by applying the Ångström equation (Ångström, 1929) to match the MODIS AOD_{550nm} . For these intense DD episodes, the comparison between the satellite and ground aerosol optical depths at 550 nm is given in Figure 5. Two similar scatterplots with matched MODIS-AERONET data pairs are given. The first one (Fig. 5 i-a) is resolved by the number of level 2 (L2) measurements of 10 km x 10 km spatial resolution from which the

533 compared $1^\circ \times 1^\circ$ level 3 (L3) *AODs* in the figure are derived. The second scatterplot (Fig. 5 i-b) is
534 resolved by the spatial standard deviation inside the $1^\circ \times 1^\circ$ geographical cell (level 3 *AODs*). Both
535 scatterplots address the issue of level 3 *AOD* sub-grid spatial variability, which is essential when
536 attempting comparisons against local surface-based *AOD* data like the AERONET.

537 The overall correlation coefficient (*R*) between MODIS and AERONET *AODs* is equal to 0.505,
538 with the satellite *AODs* being overestimated (bias=0.143). From the overall scatterplots, it is evident
539 the existence of outliers associated with small number of level 2 retrievals (<20, blue color Fig. 5 i-a)
540 and/or high standard deviations (>0.5, yellowish-reddish points, Fig. 5 i-b) inside the L3 grid cell. This
541 finding underlines the role of homogeneity and representativeness of L3 retrievals for the comparison
542 of MODIS *AODs* against AERONET. This role is better visualized in Fig. 5 ii-a, where are presented
543 the computed *R* values between MODIS level-3 and AERONET *AODs* depending on the number of L2
544 retrievals from which the L3 products were derived. In general, it is known that the L2 pixel counts
545 range from 0 to 121 while in polar regions (typically around 82° latitude) the maximum count numbers
546 can be even higher due to overlapping orbits and near nadir views intersect (Hubanks et al., 2008). It is
547 clear from our results that the correlation coefficients are gradually and essentially improved, from 0.49
548 to 0.75, with increasing representativeness of MODIS *AODs*, i.e. increasing counts of L2 retrievals
549 attributed. A similar improvement has been reported by Amiridis et al. (2013) who found a better
550 agreement between MODIS/AERONET and CALIOP aerosol optical depths applying similar spatial
551 criteria. The agreement between MODIS and AERONET also improves when the former *AOD*
552 products are more spatially homogeneous, i.e. when they are characterized by smaller *AOD* standard
553 deviations at the grid-level (from <0.25 down to <0.05, Fig. 5 ii-b). However, our results also indicate
554 that apart from increasing correlation coefficients (up to 0.7-0.8) with increasing level-2 counts and
555 decreasing standard deviations, the number of intense DD episodes is decreased dramatically (about
556 40-50 for more than 50 counts and standard deviation smaller than 0.05).

557 In addition, the spectral variation of the AERONET *AODs* at 7 wavelengths, from 340 to 1020 nm,
558 in climatological and dust episodes conditions has been investigated (results given in Figure S3,
559 supplementary material). The *AOD* boxplots produced for all the available daily AERONET
560 measurements (orange) and for the corresponding retrievals during strong (cyan), extreme (red) and all
561 DD (green) episodes identified by the satellite algorithm show that the spectral variation of aerosol
562 optical depth decreases in cases of dust episodes, with respect to the “climatological” conditions. This
563 is mainly attributed to the further increasing *AOD* levels at wavelengths longer than 500 nm (by about
564 6 times) than in (or near) the visible.

566 *4.2.1.2 Aerosol volume size distribution*

567 In Figure 6, are presented the mean aerosol volume size distributions (AVSDs) calculated from all
 568 available AERONET data (orange curve) as well as under strong (cyan curve), extreme (red curve) and
 569 all (green curve) DD episodes conditions. The results are given for Mar. 2000 – Feb. 2013 using
 570 MODIS-Terra (346 intense DD episodes) retrievals as inputs to the satellite algorithm. In the
 571 climatological curve, two modes are distinct centered at 0.15 μm for the fine mode and 2.24 μm for the
 572 coarse mode. There is an about equal contribution of both modes, indicating the coexistence of fine
 573 (e.g. urban aerosols) and coarse (e.g. dust aerosols) particles over the broader Mediterranean area. This
 574 result is in agreement with previous studies for the Mediterranean (e.g. Fotiadi et al., 2006; Mallet et
 575 al., 2013). However, under dust episode conditions, although the AVSD still has two modes, there is a
 576 dramatic increase of the coarse mode, which strongly dominates. More specifically, the peak of the
 577 coarse mode (radius between 1.7 and 2.24 μm) is increased by factors of about 10, 15 and 11 for the
 578 strong, extreme and all DD episodes. The differences between the strong and extreme AVSDs are
 579 statistically significant (confidence level at 95 %) for almost all size bins (18 out of 22) except bin 1
 580 (0.050 μm), 2 (0.065 μm), 6 (0.194 μm) and 7 (0.255 μm). Moreover, it should be noted that the
 581 increment factors are slightly decreased when the algorithm operates only with AODs associated with
 582 cloud fractions less than 0.8 which is reasonable since possible “overestimated” retrievals are masked
 583 out from the analysis. Similar modifications in the shape of AVSD during dust outbreaks have been
 584 pointed out by several studies in the past, either for the Mediterranean region (e.g. Kubilay et al., 2003;
 585 Lyamani et al., 2005; Córdoba-Jabonero et al., 2011) or for other dust affected areas of the planet (e.g.
 586 Alam et al., 2014; Cao et al., 2014).

587 *4.2.1.3 Size optical properties, single scattering albedo and asymmetry parameter*

588 The accuracy of the DD episodes identification method was further assessed by also using other
 589 AERONET aerosol optical properties than AOD, namely the Ångström exponent (α) and the effective
 590 radius (r_{eff}), able to provide information about particles' size. For both aerosol optical properties, the
 591 boxplots for all the available AERONET retrievals as well as for the corresponding data during strong,
 592 extreme and all DD episodes, have been produced and depicted in Figure S4 (supplementary material).

593 Based on our results, the appropriateness of the applied methodology is confirmed by the drastic
 594 reduction of α and increase of r_{eff} values when dust outbreaks occur. When all available AERONET
 595 retrievals are considered (clim), the majority (>75%) of α values is higher than 1.04 indicating the

strong presence of fine particles in the study domain. On the contrary, during intense dust episodes the 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_{eff} values under intense DD conditions compared to the climatological levels. For all DD episodes, the 75% of r_{eff} values is higher than 0.55 μm reaching up to 1.4 μm , 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 S5-i) and g_{aer} (Figure S5-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 values, by up to 0.04 and 0.07, 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.

4.2.1.4 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 4, 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

627 Middle East deserts) and (iii) the inclusion of sites where the aerosols' regime is complex (e.g. El
628 Arenosillo, FORTH Crete). The intense DD episodes were identified following the methodology
629 described in section 3, but using only AOD at 870 nm, $\alpha_{440-870nm}$ (lower/equal than 0.7) and r_{eff} (higher
630 than 0.6) as criteria, based upon their availability from AERONET. Subsequently, the algorithm was
631 also operated again using satellite (MODIS-Terra, OMI-Aura, EP-TOMS) input data for the days with
632 available retrievals in each of the 7 AERONET stations.

633 In Figure 7, we present the overall scatterplots between satellite and ground $AODs$ when intense
634 DD episodes have been identified based on the ground (left column) and the satellite (right column)
635 algorithm. Colors in Figs. 7 i-a, 7 ii-a, 7 iii-a represent the associated MODIS-Terra Ångström
636 exponent, effective radius and day cloud fraction (CFD) retrievals, respectively. In Figs. 7 i-b and 7 ii-b
637 colors represent the AERONET Ångström exponent and effective radius, respectively, while in Figure
638 7 iii-b represent the day cloud fraction observations derived by MODIS-Terra. Through this approach it
639 is feasible to assess furthermore the performance of the satellite algorithm, specify its drawbacks and
640 check the validity of the defined thresholds (green boxes in Figure 1).

641 It is apparent that the agreement between MODIS-Terra and AERONET $AODs$ is better when DD
642 episodes are identified from the ground, as shown by the increased correlation coefficients (from 0.521
643 to 0.704), increased slopes (from 0.6 to 0.9-1.0) and decreased biases (from 0.16 to -0.03). In
644 particular, when DD episodes are identified from space, the MODIS-Terra AOD retrievals are
645 overestimated (bias=0.163) with regards to AERONET, particularly at low AOD values (<0.5). In both
646 algorithms, the highest overestimations are associated with cloud fractions higher than 0.7 due to the
647 possible contamination of the satellite $AODs$ by clouds (Figure 7 iii-a, iii-b). Given that DD episodes'
648 identification based on AERONET retrievals is more efficient, we have used these results in order to
649 check the validity of the defined thresholds for α , AI , FF and r_{eff} (green boxes in Figure 1) used in the
650 satellite algorithm. For each aerosol optical property, it has been calculated the percentage of intense
651 DD episodes for which the corresponding satellite observations are below or above the defined
652 thresholds, depending on the parameter. The results given in Table 2 are satisfactory, since the
653 percentages range from 87 to 99%, and confirm the validity of the defined thresholds.

654 The scatterplots in Figs. 7 i-b and ii-b also reveal some weaknesses of the satellite-based algorithm.
655 More specifically, it is found that for few DD episodes identified by the satellite algorithm the
656 corresponding AERONET Ångström exponent and effective radius values are higher than 1 and
657 smaller than 0.4, respectively. These values indicate a predominance of fine particles instead of coarse

ones as it would be expected for desert dust aerosols. In order to quantify the number of misclassified pixel level intense DD episodes by the satellite algorithm, we have computed the percentage of cases for which the AERONET α values are higher than 1 (15%) and r_{eff} values are lower than 0.4 (17.7%). Also, we have repeated these calculations for all DD episodes (Section 4.2.1.1) and the corresponding percentages were found to be equal to 11.8% and 14.5%, respectively. These misclassifications of the satellite algorithm occur in AERONET stations (e.g. Thessaloniki, Rome, Avignon) with a strong presence of anthropogenic aerosols (Kazadzis et al., 2007; Gobbi et al., 2007; Querol et al., 2009a; Yoon et al., 2012). Some misclassifications also occur in AERONET stations (e.g. Evora, El Arenosillo, FORTH CRETE) with mixed (natural plus anthropogenic) aerosol loads (Fotiadi et al., 2006; Toledano et al., 2007b; Hatzianastassiou et al., 2009; Pereira et al., 2011). Over these areas, there are converging air masses carrying particles of different origin, as shown by performed back-trajectories analyses (results are not shown here) using the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph, 2015). Nevertheless, it must be mentioned that DD episodes' misclassifications can be attributed to the lower accuracy of MODIS aerosol size retrievals over land (Section 2.1.1).

673

674 4.2.2 PM₁₀ and dust contribution

675 The satellite algorithm's outputs, apart from AERONET retrievals, have been also compared
676 against ground PM₁₀ concentrations ($\mu\text{g m}^{-3}$) measured in 22 Mediterranean stations (green triangles in
677 Figure 4).

678 First, for each station, the number of intense DD episodes was calculated, for which coincident
679 satellite and ground measurements (total PM₁₀) are available (Figure 8-i). The number of concurrent
680 DD episodes varies from 3 to 53, being in general decreasing from southern to northern stations. For 14
681 out of 22 stations, where at least 10 intense DD episodes were identified by the satellite-based
682 algorithm, we have computed the correlation coefficients between satellite *AODs* and surface total
683 PM₁₀ concentrations (Fig. 8-ii). The highest R values (up to 0.8) are recorded in the central and eastern
684 parts of the Mediterranean while the lowest ones are found in the western stations. It must be noted that
685 the correlation coefficients are affected by outliers, because of the limited number of DD episodes in
686 each station, highlighting the sensitiveness of the intercomparison. Such outliers can be expected when
687 satellite-based columnar *AODs* and surface-based PM₁₀ data are compared, since satellite *AODs* are
688 representative for the whole atmospheric column in contrast to in-situ PM measurements which are

689 more representative for the lowest part of the planetary boundary layer affected also by local factors.
690 Therefore, the vertical distribution of desert dust load, as it will be presented in the next sections, can
691 determine the level of agreement between satellite *AODs* and surface PM concentrations. Another
692 influencing factor can be cloud contamination of MODIS *AOD*.

693 The identification method by the satellite algorithm can be considered as correct when dust PM₁₀
694 concentrations are higher than zero (i.e. dust has been recorded at the station). According to this, the
695 ratio between the number of non-zero dust PM observations and the number of DD episodes
696 (coincident satellite-derived DD episodes and total PM₁₀ measurements) for each station is defined as
697 success score. The calculated success scores (Figure 8-iii) vary from 68% (Monagrega, northeastern
698 Spain, 28 episodes) to 97% (Boccadifalco, Sicily, 33 episodes) confirming the appropriateness of the
699 DD episodes' identification. In the majority of stations, the contribution of dust particles to the total
700 burden (Figure 8-iv) is above 50%, ranging from 44% (Zarra, Spain) to 86.8% (Ayia Marina, Cyprus).
701 In order to complete our analysis we have also calculated the mean (Figure 8-v) and the median (Figure
702 8-vi) dust PM₁₀ concentrations for the identified intense DD episodes in each station. The mean PM₁₀
703 concentrations mainly vary between 20 and 50 $\mu\text{g m}^{-3}$, being higher in the southern stations, as
704 expected. The minimum mean value (17 $\mu\text{g m}^{-3}$) was recorded in Censt (Sardinia) and the maximum
705 one (223 $\mu\text{g m}^{-3}$) in Ayia Marina (Cyprus). Our values are much higher than the corresponding ones in
706 Querol et al. (2009b), who obtained that the mean levels of mineral matter in PM₁₀ during dusty days
707 range from 8 to 23 $\mu\text{g m}^{-3}$ based on ground concentrations derived by 21 Mediterranean stations. These
708 differences are reasonable since here only intense desert dust outbreaks associated with high aerosol
709 optical depths are considered. Finally, the median PM₁₀ concentrations are lower compared to the
710 average ones, indicating that outliers (cases with extremely high *AOD* or PM₁₀) can alter the results,
711 attributed to the fact that both parameters' (*AOD* and PM₁₀) distributions are not Gaussians. For this
712 reason the highest differences are found in Finokalia (Crete) and Agia Marina (Cyprus), where the
713 maximum daily PM₁₀ concentrations, equal to 690 and 1291 $\mu\text{g m}^{-3}$, respectively, were recorded during
714 an intense dust outbreak affected the eastern Mediterranean on 24 and 25 February 2006.

715

716 4.3 Vertical structure of the Mediterranean desert dust outbreaks

717 The ability of the developed satellite algorithm to detect intense dust episodes has been proved
718 adequate through the comparison analysis against AERONET retrievals and PM₁₀ concentrations.
719 Nevertheless, its main limitation is that it uses columnar satellite retrievals and not vertical resolved

720 data prohibiting thus the description of the vertical structure of these dust outbreaks. In order to address
721 this issue, the CALIOP-CALIPSO retrievals are used as a complementary tool to the satellite
722 algorithm's outputs. First, for the identified dust episodes by the satellite algorithm, the spatially and
723 temporally collocated vertically resolved CALIOP lidar observations are selected. For these cases and
724 for each $1^\circ \times 1^\circ$ grid cell, we have divided the lower troposphere, up to 8 km, in 16 layers of 500
725 meters height. In this way, 14400 boxes of $1^\circ \times 1^\circ$ surface area and 500 meters height have been
726 produced. Then, for each one of them, we have calculated the overall number of dust and polluted dust
727 observations (hereafter named as dust) according to the aerosol subtyping scheme of the CALIOP
728 Vertical Feature Mask (VFM). Note that dust and polluted dust were chosen because in previous
729 studies (Mielonen et al., 2009) they were shown to be the best two defined aerosol types among the
730 other ones classified by the CALIOP VFM. Nevertheless, in case of polluted dust, Burton et al. (2013)
731 reported that dust particles can be mixed with marine aerosols instead of smoke or pollution as assumed
732 by the VFM retrieval algorithm. In our study, more than 95% of the aerosol type records were pure
733 dust, for the collocated cases between the satellite algorithm and CALIPSO observations. In addition,
734 in the majority of the defined boxes, the percentage of dust from the overall observations is higher than
735 70%, confirming furthermore the validity of the algorithm DD episodes' identification procedure. This
736 is an excellent proof of the successful identification of DD episodes by the satellite algorithm, since
737 CALIOP-CALIPSO is an independent and vertically resolved platform and database. Thereby,
738 CALIOP vertical observations were subsequently used to examine the vertical structure of dust
739 outbreaks.

740 In order to analyze the intensity of desert dust outbreaks at different altitudes in the troposphere, the
741 CALIOP data of the total backscatter coefficient at 532 nm (β_{532nm}) have been also acquired. For each
742 box, the average β_{532nm} values have been calculated from all the available CALIOP measurements (day
743 and night), for the identified intense dust episodes by the satellite algorithm. More specifically, the
744 average β_{532nm} values were calculated for the dust observations based on the CALIOP VFM associated
745 with CAD scores ranging from -100 to -20, as it has been proposed by Winker et al. (2013) for
746 discriminating aerosol from clouds. The selection of β_{532nm} values instead of extinction coefficients
747 ensures that incorrect lidar ratio assumptions in the CALIOP retrieval algorithm do not affect our
748 results. In the literature, it has been documented that the CALIOP lidar ratio is underestimated over the
749 northern African deserts and the surrounding areas affected by Saharan dust particles, leading to an
750 underestimation of the columnar *AOD* compared to MODIS and AERONET retrievals (Redemann et
751 al., 2012; Schuster et al., 2012). Amiridis et al. (2013) stated that an increase of the lidar ratio from 40

752 to 58 sr, along with a series of post-corrections in the CALIOP retrievals and the implementation of
753 several criteria concerning the cloud coverage and the spatial representativeness, can improve
754 substantially the agreement between MODIS-Aqua/AERONET and CALIOP observations.

755 It should be noted that in the present work, we have analyzed all the available CALIOP overpasses
756 (~ 10000) over the study region, during the period Jun. 2006 – Feb. 2013. For brevity reasons,
757 however, only the obtained results based on MODIS-Terra retrievals are presented here, since similar
758 findings are drawn for MODIS-Aqua (Jun. 2006 – Dec. 2012). Moreover, the analysis (results are not
759 shown here) has been made separately for the identified strong and extreme DD episodes without
760 revealing remarkable differences in the geometrical characteristics of dust outbreaks. Nevertheless, the
761 β_{532nm} values are higher for the extreme DD episodes being consistent with the discrimination of dust
762 episodes' intensity (in terms of *AOD*) which is applied to the satellite algorithm. In order to facilitate
763 the visualization of our results, for each column (1° x 1° spatial resolution) and latitudinal/longitudinal
764 zone (1° degree), we have calculated the overall number of dust observations and the associated
765 weighted averages of β_{532nm} , depending on the projection plane (latitudinal, longitudinal and columnar),
766 according to dust observations in each box. For both parameters, the analysis has been made on an
767 annual and seasonal basis and the corresponding results are discussed in Sections 4.3.1 and 4.3.2,
768 respectively.

769

770 4.3.1 Annual characteristics

771 In Figure 9, are presented the three dimensional structures of the CALIOP overall dust observations
772 (Fig. 9-i) and the associated total backscatter coefficients at 532 nm (Fig. 9-ii), during intense dust
773 episodes conditions, over the broader Mediterranean area, for the period Jun. 2006 – Feb. 2013. From
774 the latitudinal projection in Fig. 9-i, it is evident that dust particles are mainly detected between 0.5 and
775 6 km, and more rarely up to 8 km, between the parallels 32° N and 38° N. The number of dust
776 observations is increased at higher altitudes with increasing latitudes, up to 40° N, while the altitude
777 range (thickness) where these records are detected is gradually reduced from 4 to 2 km. At northern
778 latitudes, the CALIPSO dust records are drastically reduced and are mainly observed between 1 and 4
779 km. The ascending mode of the transported mineral particles over the Mediterranean is attributed to the
780 prevailing low pressure systems, which mobilize and uplift dust particles from the source areas across
781 the Sahara Desert and the Arabian Peninsula. Dust aerosols are transported over the planetary boundary
782 layer (Hamonou et al., 1999) due to the upward movement of dry and turbid air masses (Dulac et al.,

1992), while the prevailing synoptic conditions determine also the spatial and temporal characteristics of desert dust outbreaks over the Mediterranean (Gkikas et al., 2014).

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

As to the variation of vertical extension with longitude (Fig. 9-i), it is revealed that the base height of dust layers is decreased towards the eastern parts of the study region. In the western Mediterranean, the mineral particles are mainly detected between 2 and 6 km while over the central and eastern Mediterranean the corresponding altitudes are equal to 0.5 and 6 km, respectively. It is well known, that dust is transported over the western Mediterranean mainly in summer (e.g. Moulin et al., 1998) favored by low pressure systems located over the northwestern Africa (Gkikas et al., 2014) and the enhanced thermal convection, uplifting effectively dust aerosols at high altitudes in the troposphere. Moreover, air masses carrying dust particles are “convected” towards higher altitudes due to the existence of the Atlas Mountains Range. Therefore, the combination of strong convective processes over North Africa along with topography can explain the identification of dust aerosols at higher tropospheric levels over the western Mediterranean. It is the presence of mineral particles at high altitudes in western Mediterranean that can explain the poor-to-moderate agreement between PM_{10} concentrations and MODIS *AODs* found in the Iberian Peninsula (Fig. 8-ii). In order to give a better insight to how the dust outbreaks’ vertical extension can affect the level of agreement between columnar AOD satellite retrievals and ground PM_{10} concentrations, emphasis is given at specific dust events and the relevant findings will be discussed in section 4.4. In the central and eastern parts of the Mediterranean basin, air masses carrying African dust aerosols travel at lower altitudes over Africa because of absence of significant topographical objects on their route, as suggested by Pey et al. (2013).

815 Previous studies have shown that dust layers over the Mediterranean are characterized by a
816 multilayered structure (e.g. Hamonou et al., 1999; Mona et al., 2006; Papayannis et al., 2008). This is
817 also depicted in the longitudinal projection of Figure 9-i, where several dust layers of different base/top
818 altitudes and geometrical thicknesses are detected. In general, the base heights vary from 0.5 to 2 km,
819 the top heights from 4 to 6 km and the thicknesses from 1 to 4 km. The majority of common
820 observations between the CALIOP profiles and the identified intense DD episodes by the satellite
821 algorithm are recorded over the maritime parts of the study region (bottom map of Fig. 9-i). The
822 maximum number of CALIOP dust observations (~ 19000) is recorded along the Atlantic coasts of
823 Morocco, but high numbers (about 10000 – 15000) are also found across the northern African coasts.

824 Apart from the CALIOP dust observations, we have also analyzed the associated β_{532nm} values at
825 the defined altitude ranges in order to describe the variation of intensity of the desert dust episodes with
826 height over the Mediterranean (Fig. 9-ii). The maximum backscatter coefficients (up to $0.006 \text{ km}^{-1} \text{ sr}^{-1}$)
827 are observed below 2 km, being increased towards the southern edges ($30^\circ \text{ N} - 34^\circ \text{ N}$) of the study
828 region, where their source areas are found. This is explained by the fact that dust particles due to their
829 coarse size and large mass, are efficiently deposited and for this reason they are recorded at higher
830 concentrations near to the source areas and at low altitudes. Nevertheless, the decreasing intensity with
831 height towards the north is not so evident. Thus, high β_{532nm} values ($\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$) are observed
832 between 2 and 4 km in the latitudinal zone extending from 35° N to 42° N . Though, the uppermost
833 altitudes where relatively high β_{532nm} values gradually decrease from 6 to 4 km, moving from south to
834 north. Any differences in the latitudinal patterns of dust observations and backscatter values (Figs 9-i
835 and 9-ii) can be explained by the fact that β_{532nm} values take into account only the dust records and not
836 the overall observations (all aerosol types).

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

846 The longitudinal pattern of β_{532nm} profiles (Fig. 9-ii) is less distinct compared to the corresponding
847 one resulted from the latitudinal projection. Relatively high β_{532nm} values ($\sim 0.004 \text{ km}^{-1} \text{ sr}^{-1}$) are found
848 between 1 and 5 km over the western Mediterranean, while over the central and eastern parts of the
849 study region the desert dust outbreaks' intensity ($\sim 0.006 \text{ km}^{-1} \text{ sr}^{-1}$) is higher below 1.5 km. Among the
850 sub-regions, the backscatter coefficients are higher in the central and eastern Mediterranean, which is
851 also depicted in the bottom map of Fig. 9-ii. It is reminded that higher intensities of dust episodes over
852 the central and eastern Mediterranean have also been noticed based on MODIS retrievals (Figure 3).
853 From the obtained longitudinal projection, it is evident a patchy structure of the total backscatter
854 coefficient profiles, especially in the central and eastern parts, indicating the existence of several dust
855 layers of varying intensities at different altitudes into the atmosphere.

856 The three dimensional plots of Figures 9-i and 9-ii, have been also reproduced considering all the
857 available dust and polluted dust CALIOP-CALIPSO records, without taking into account the satellite
858 algorithm's outputs (for intense dust outbreaks). The obtained results for the number of observations
859 and β_{532nm} are presented in Figures S6-i and S6-ii, respectively. Note, that for each studied parameter
860 the colorbar scales in Figure 9 and S6 are not identical because the number of observations for dust
861 average conditions (Fig. 6-i) is extremely larger than the corresponding one during intense dust
862 outbreaks (Fig. 9-i) while the opposite is found for the β_{532nm} values (Fig. 9-ii and Fig. 6-i). It is
863 apparent that the latitudinal projections calculated for the intense dust outbreaks (Fig. 9-i) and for all
864 the available CALIOP dust records (Fig. S6-i) reveal different patterns. More specifically, when all
865 available CALIOP dust records are considered, it is found that dust aerosols are mainly confined
866 between 1 and 3 km in the southernmost parts of the study region while the number of observations
867 gradually decreases at higher altitudes and towards northern latitudes (Fig. S6-i). On the contrary,
868 during dust outbreaks, mineral particles are transported over the Mediterranean following an ascending
869 path, as it is depicted in the latitudinal projection of Figure 9-i. Nevertheless, it must be mentioned that
870 over the desert areas there is a full coverage (see bottom map in Fig. S6-i) when all dust CALIOP
871 records are considered in contrast to intense dust outbreaks (see bottom map in Fig. 9-i) attributed to
872 the absence of DT retrievals, used as inputs to the satellite algorithm, over bright surfaces. The
873 comparison between the longitudinal projections during intense dust outbreaks (Figure 9-i) and during
874 average dust conditions (Fig. S6-i) reveals less remarkable differences than for the latitudinal
875 projections. According to the longitudinal projection of Figure S6-i, in the western Mediterranean, dust
876 layers are confined between 1 and 6 km, while their base and top altitude both decrease down to 0.5
877 and 4.5 km, respectively, for increasing longitudes. In the easternmost part of the study region, dust

878 layers are mainly confined between 1 and 3 km, while its top height can reach up to 5 km. The intensity
879 of dust loads (in terms of β_{532nm}) is lower than $0.003 \text{ km}^{-1} \text{ sr}^{-1}$ regardless the projection plane for
880 average dust conditions based on CALIOP-CALIPSO lidar profiles (Fig. S6-ii). Moreover, the intensity
881 of dust loads decreases gradually with height as well as from south to north revealing a distinct pattern
882 in all projection planes in contrast to the corresponding ones found during desert dust outbreaks (Fig. 9-
883 ii).

884

885 4.3.2 Seasonal characteristics

886 The vertical structure of the Mediterranean desert dust outbreaks has also been analyzed separately
887 for winter (DJF), spring (MAM), summer (JJA) and autumn (SON). The seasonal three dimensional
888 representations of the CALIOP overall dust observations and the associated total backscatter
889 coefficients are depicted in the left and right column of Figure 10, respectively. It must be noted, that
890 for β_{532nm} the colorbars' ranges are common, depending on the projection plane. More specifically, the
891 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,
892 longitudinal and bottom map projections, respectively. It should be mentioned that β_{532nm} values can
893 reach up to $0.045 \text{ km}^{-1} \text{ sr}^{-1}$, but are associated with a very small number of dust observations.

894 The majority (85%) of dust observations is recorded in spring and summer, attributed to the
895 enhanced production rates of mineral particles and the prevailing atmospheric circulation over the
896 source areas and the Mediterranean. According to the latitudinal projections, it is evident a seasonal
897 variability of the intense Mediterranean desert dust outbreaks' geometrical characteristics. Dust
898 particles are detected at higher altitudes (6-7 km) during warm seasons of the year while in winter are
899 mainly detected below 3 km and in autumn are recorded between 2 and 5 km. Nevertheless, it should
900 be mentioned that during these seasons only a small number of pixels (see bottom maps in Figs. 10 i-a,
901 iv-a) is available considering also that clouds prohibit the satellite observations. Note that in spring,
902 dust can be found at low tropospheric levels while in summer it is mainly observed above 1 km
903 highlighting thus the role of topography and the enhanced thermal convection. During the first half of
904 the year, the maximum dust observations are confined between the parallels 31° N and 37° N while
905 during the second one, are shifted northwards in the latitudinal zone extending from 34° N to 40° N .
906 Similar latitudinal projections were also presented by Luo et al. (2015), for the same zonal areas of the
907 study region, who developed a new algorithm to improve CALIOP's ability to detect optically thin dust
908 layers. From the longitudinal projections as well as from the bottom maps, it is evident that the

909 maximum dust records are found in different Mediterranean sub-regions, depending on the season. The
910 geometrical characteristics, in longitudinal terms, of intense DD episodes affecting the western, central
911 and eastern parts of the Mediterranean are similar to those presented in the annual three dimensional
912 structure (Fig. 9-i) being more frequent in the eastern and central Mediterranean in winter, spring and
913 autumn and in the western and central Mediterranean in summer.

914 The seasonal patterns of β_{532nm} latitudinal projections are different than those for the dust
915 observations, while they also differ among the four seasons. The intensity of winter DD episodes is
916 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
917 the longitudinal and bottom map projections, these episodes take place over the central and eastern
918 Mediterranean Sea but the number of grid cells with coincident CALIOP observations and DD episodes
919 is limited. In spring, the highest β_{532nm} values (up to $0.006 \text{ km}^{-1} \text{ sr}^{-1}$) are recorded between the parallels
920 31° N and 35° N and below 2 km, although, relatively high β_{532nm} values (up to $0.004 \text{ km}^{-1} \text{ sr}^{-1}$) are
921 found up to 6 km (Fig. 10 ii-b). Moving northwards, over the Mediterranean, dust layers are mainly
922 confined between 2 and 4 km, associated with high β_{532nm} values (up to $0.004 \text{ km}^{-1} \text{ sr}^{-1}$) in the
923 latitudinal zone extending from 35° N to 43° N . The existence of these elevated dust layers, has been
924 also confirmed by model simulations through specific (Papayannis et al., 2008; 2014) or averaged
925 (Alpert et al., 2004) cross sections of dust concentrations in the central sector of the Mediterranean.
926 This is in accordance with our longitudinal projection (Fig. 10 ii-b), where β_{532nm} is high varying from
927 0.004 to $0.008 \text{ km}^{-1} \text{ sr}^{-1}$ at these altitude ranges.

928 In summer, the intensity of dust episodes is smoothly decreased at higher altitudes, where dust
929 layers of considerable β_{532nm} values are also found. More specifically, the highest backscatter
930 coefficients (up to $0.008 \text{ km}^{-1} \text{ sr}^{-1}$) are recorded near to the surface but also moderate values (up to
931 $0.006 \text{ km}^{-1} \text{ sr}^{-1}$) are observed between 2 and 5 km, particularly over the southern parts of the study
932 region (Fig. 10 iii-b). Most of these intense DD episodes occur in the western Mediterranean, where the
933 highest β_{532nm} values (up to $0.005 \text{ km}^{-1} \text{ sr}^{-1}$) are recorded between 2 and 5 km. Over the central and
934 eastern Mediterranean, even higher β_{532nm} values are found (up to $0.014 \text{ km}^{-1} \text{ sr}^{-1}$) but at lower altitudes
935 ($< 1 \text{ km}$). In autumn, the majority of the grid cells of coincident CALIOP profiles and DD episodes
936 identified by the satellite algorithm are located between the parallels 33° N and 41° N . In this
937 latitudinal zone, CALIOP profiles are available over the interior parts of the Iberian Peninsula and over
938 western and central parts of the Mediterranean Sea, near to the northern African coasts. According to
939 the latitudinal projection, β_{532nm} values mainly vary from 0.002 to $0.009 \text{ km}^{-1} \text{ sr}^{-1}$, revealing an
940 increasing tendency for increasing heights. On the contrary, the total backscatter coefficients do not

941 show a distinct spatial pattern on the longitudinal projection, due to the limited number of grid cells
942 participating in the calculations. Throughout the year, based on the CALIOP β_{532nm} retrievals, the DD
943 episodes are more intense (up to $0.018 \text{ km}^{-1} \text{ sr}^{-1}$) in spring, when massive dust loads are transported
944 from the Sahara desert towards the central and eastern parts of the Mediterranean Sea (bottom map in
945 Fig. 10 ii-b).

946

947 4.4. Intercomparison of satellite AOD and PM₁₀ concentrations for specific desert dust outbreaks

948 In Section 4.2.2, it has been shown that the agreement between the satellite algorithm's outputs and
949 PM₁₀ concentrations is better in the central and eastern Mediterranean with regards to the western parts
950 (Figure 8-ii). This discrepancy has been mainly attributed to the higher altitude of dust layers' base
951 over the western sector of the study domain (Figure 9-i), in relation to the existing areal orography.
952 Here, aiming at addressing how dust layers' geometrical characteristics influence the agreement
953 between columnar AOD satellite and ground PM₁₀ measurements, specific desert dust outbreaks that
954 took place over the PM₁₀ stations are analyzed. These outbreaks were selected based on concurrent
955 fulfillment of the following criteria: (i) a DD episode must be identified by the satellite algorithm at
956 pixel level (at $1^\circ \times 1^\circ$ grid cell), (ii) total PM₁₀ measurement must be available at the station which lies
957 into the geographical limits of the corresponding grid cell and (iii) CALIPSO flies across the grid cell.
958 These criteria were met for 13 desert dust outbreaks, which took place over 9 PM₁₀ stations during the
959 period 2000-2013. Similarities were found among the identified cases and therefore only the results for
960 four desert dust outbreaks of different geometrical characteristics are discussed in the present section.
961 For each case, we have produced the cross sections of the β_{532nm} vertical profiles up to 8 km above sea
962 level (a.s.l.) along the CALIOP-CALIPSO track when the satellite flies near the PM₁₀ site (Figures 11-
963 13). Moreover, the corresponding aerosol subtype profiles, acquired from the CALIOP website
964 (http://www-calipso.larc.nasa.gov/products/lidar/browse_images/production/), are provided in the
965 supplementary material (Figures S7-S9). Since the PM₁₀ concentrations are available only as daily
966 averages, the optimum solution would be to have the maximum number (2) of CALIOP overpasses
967 near PM₁₀ site throughout the day, in order to reduce the temporal inconsistencies between satellite
968 vertical resolved retrievals and ground data. However, in 8 out of 13 desert dust outbreaks this was not
969 feasible.

970

971 4.4.1 Case 1: Censt (26th May 2008)

972 The first study case refers to a desert dust outbreak that took place on 26th May 2008 and affected
973 the station Censt (Lat: 39.064, Lon: 8.457) located in southern Sardinia. At the ground, the measured
974 mean daily total PM₁₀ concentration was 19 µg m⁻³ whereas 68% (or 13 µg m⁻³) of the load consisted of
975 dust particles indicating thus their strong presence in the lowest troposphere. Based on MODIS-Terra
976 retrievals, representative for the whole atmospheric column and grid cell, the aerosol optical depth at
977 550 nm was equal to 0.81. In order to investigate the vertical distribution of the dust outbreak, the cross
978 sections of the β_{532nm} vertical profiles along CALIOP track, near the station, during daytime and
979 nighttime have been reproduced and depicted in Figures 11-i and 11-ii, respectively. In addition, the
980 corresponding aerosol subtype profiles are provided in Figures S7-i and S7-ii in the supplementary
981 material. During night, it is evident the predominance of a well-developed dust layer mixed with
982 polluted aerosols (Figure S7-i) extending from surface up to 5 km a.s.l. between the parallels 33° N and
983 38° N, while near the station its top is lowered down to 3 km (left side of Figure 11-i). Moreover, the
984 β_{532nm} values range mainly from 0.002 to 0.003 km⁻¹ sr⁻¹ without revealing remarkable variations, thus
985 indicating a rather compact dust layer. According to the daytime CALIOP overpass (Figure 11-ii), a
986 pure dust layer (Figure S7-ii) is confined between surface and 4 km, affecting the surrounding area of
987 the station, while its intensity (in terms of β_{532nm}) varies slightly from 0.0015 to 0.002 km⁻¹ sr⁻¹.
988 Nevertheless, due to the background solar illumination, leading thus to a lower signal-to-noise ratio
989 (Nowottnick et al., 2015), the “borders” of the dust plume during daytime are not so distinct in contrast
990 to nighttime. According to the obtained results, the ground-based measurements are able to capture
991 satisfactorily the dust event when its load is equally distributed in the lowest tropospheric levels,
992 resulting thus to a good agreement between MODIS and PM₁₀ observations.

993

994 4.4.2 Case 2 and 3: Els Torms (16th July 2008) and San Pablo (12th September 2007)

995 Two dust events that affected Els Torms (NE Spain, Lat: 41.395, Lon: 0.721) and San Pablo
996 (central Spain, Lat: 39.525, Lon: -4.353) on 16th July 2008 and 12th September 2007, respectively, are
997 studied here. The daily averages of the total PM₁₀ concentrations were equal to 16 and 30 µg m⁻³,
998 respectively, whereas the dust particles' contribution (dust PM₁₀) to the total amount was zero in Els
999 Torms and 33 % in San Pablo. On the contrary, the MODIS-Terra level 3 AOD retrievals were high
1000 and equal to 0.56 (Els Torms) and 0.64 (San Pablo), indicating the existence of dust aerosols according
1001 to the satellite algorithm's classification method. In order to give a better insight, aiming at describing

the discrepancies between MODIS-Terra AOD and PM₁₀ concentrations, we have reproduced the cross sections of the total backscatter at 532 nm when CALIPSO flies, during daytime, near Els Torms (Figure 12-i) and San Pablo (Figure 12-ii). The corresponding profiles of the CALIOP aerosol classification scheme are also available in Figures S8-i and S8-ii. In Els Torms, where the dust PM₁₀ concentration was zero, a dust layer (Figure S8-i) with its base at 3.5 km a.s.l. and its top at 5 km a.s.l., is recorded by the CALIOP lidar between the parallels 41° N and 43° N. The intensity of the elevated dust layer, in terms of β_{532nm} , varies from 0.002 to 0.004 km⁻¹ sr⁻¹ (Figure 12-i). Through CALIOP lidar profiles, it is confirmed the existence of a dust layer aloft, which cannot be captured by the PM₁₀ measurements in contrast to the MODIS spectroradiometer. In San Pablo, where the dust particles' contribution to the total PM₁₀ load was equal to 33 %, a dust layer abuts the ground extending up to 5-6 km ASL, whereas the dust plume covers a wide range, in latitudinal terms, from the sub-Sahel to the Celtic Sea, affecting the Iberian Peninsula (Figure S8-ii). Nevertheless, the intensity of the dust layer, over the surrounding area of the station, differs with altitude being higher between 2.5 and 5 km a.s.l. (0.004 to 0.007 km⁻¹ sr⁻¹) and lower between ground and 2 km a.s.l. (<0.003 km⁻¹ sr⁻¹), as it is depicted in the middle of Figure 12-ii. The two studied cases here differ from Case 1 (Section 4.4.1) either with regards to the position of the elevated dust layer (Els Torms) or to its vertical distribution (San Pablo), which explains the poor agreement between satellite columnar AOD retrievals (MODIS) and ground PM₁₀ concentrations.

4.4.3 Case 4: Agia Marina (25th February 2007)

The case studied here, namely the desert dust outbreak recorded in Agia Marina (Cyprus, Lat: 35.039, Lon: 33.058) on 25th February 2007, is the strongest one among the selected cases. More specifically, the daily average of the dust PM₁₀ concentration was equal to 134 µg m⁻³ accounting for the 92 % of the total PM₁₀ measured amount at the station, which is indicative of the strong predominance of dust particles in the lowest troposphere. The MODIS-Terra level 3 AOD value for the grid cell to which the station it is found, is high and equal to 1.04. According to the CALIOP aerosol classification scheme, during nighttime, a shallow low-elevated dust layer mixed with polluted or marine aerosols is heading towards the station, whereas above the PM₁₀ site (Agia Marina) extends from close to the ground up to 9 km a.s.l., comprising only pure dust aerosols (Figure S9). The main part of the dust layer, in the surrounding area of the station, is confined between 2.5 and 4 km a.s.l.

1033 where the maximum β_{532nm} values (up to $0.006 \text{ km}^{-1} \text{ sr}^{-1}$) are observed (Figure 13). Also, similar β_{532nm}
1034 values are recorded below 1 km a.s.l.; however, the dust layer is not well represented in the cross
1035 section of the CALIOP β_{532nm} vertical profiles due to the total attenuation of the lidar beam by clouds
1036 (located between 3 and 4 km a.s.l.) superimposed to the low-elevated dust layer.

1037

1038 **5. Summary and conclusions**

1039 This study aims at describing the vertical structure of intense desert dust outbreaks affecting the
1040 broader Mediterranean basin. To achieve this target, an updated version of an objective and dynamic
1041 algorithm, which has been introduced by Gkikas et al. (2009; 2013), has been applied for the
1042 identification of strong and extreme desert dust episodes, over the period Mar. 2000 – Feb. 2013. For
1043 its operation, a group of optical properties, retrieved by satellite sensors (MODIS-Terra/Aqua, EP-
1044 TOMS and OMI-Aura) on a daily basis, is used, providing information about aerosols' load, size and
1045 nature. Briefly, the satellite algorithm consists of three parts; at the first one are computed the mean
1046 *AOD* value (*Mean*) and the associated standard deviation (*Std*) for the whole study period in each grid
1047 cell of $1^\circ \times 1^\circ$ spatial resolution, at the second one the identified aerosol episodes are classified based
1048 on their intensity to strong and extreme ones. Finally, at the third part the identified aerosol episodes
1049 are categorized as desert dust episodes, separately over land and sea. Through this approach the
1050 selected dataset consists only of intense desert dust episodes since their intensity (expressed in terms of
1051 AOD_{550nm}) is higher/equal than $Mean + 2*Std$. The DD episodes have also been determined by
1052 applying an alternative second methodology (METHOD-B) which excludes dust-affected cases
1053 identified based on the criteria set concerning the aerosol size related optical properties.

1054 Based on the satellite algorithm's outputs, an overall view about the regime of Mediterranean desert
1055 dust outbreaks is presented for the periods Mar. 2000 – Feb. 2013 (MODIS-Terra) and 2003-2012
1056 (MODIS-Aqua). The main findings concerning the intense DD episodes' frequency (in terms of
1057 episodes yr^{-1}) and intensity (in terms of *AOD* at 550nm) are the following:

- 1058 ➤ Strong DD episodes occur more frequently (up to $9.9 \text{ episodes yr}^{-1}$) in the western
1059 Mediterranean while the extreme ones occur more frequently (up to $3.3 \text{ episodes yr}^{-1}$) over the
1060 central parts of the Mediterranean Sea, when the satellite algorithm operates with MODIS-Terra
1061 retrievals.

- The intensity of strong and extreme DD episodes, in AOD terms, can reach to 1.5 and 3-4, respectively, over the central and eastern parts of the Mediterranean Sea, near off the northern African coasts.
- Slightly lower frequencies and higher intensities are found for the period 2003-2012, when the satellite algorithm operates with MODIS-Aqua retrievals.
- Through the intercomparison between the two applied methodologies, it is revealed that the geographical patterns of frequency of occurrence are similar both for strong and extreme DD episodes; however, higher frequencies are found based on METHOD-B.
- Based on METHOD-B, the DD episodes' intensities are decreased whereas the geographical patterns for the strong DD episodes are not so distinct compared to the corresponding results obtained by the default version of the satellite algorithm.
- The similarity between the outputs of the algorithm using the two methodologies shows the consistency of the algorithm and the validity of its concept.

Through the comparison of the satellite algorithm against surface measurements derived from AERONET and 22 PM₁₀ stations, it is found that:

AERONET

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

PM₁₀ and dust contribution

- 1091 ➤ The agreement between surface and satellite measurements is better over the central and eastern
1092 Mediterranean stations.
- 1093 ➤ On a station level, the percentage of the intense DD episodes, for which a dust contribution to
1094 PM₁₀ surface concentration has been recorded, varies from 68% (Monagrega, northeastern
1095 Spain) to 97% (Bocadifalco, Sicily).
- 1096 ➤ In the majority of stations, dust particles contribute more than 50% of the total amount reaching
1097 up to 86.8% (Ayia Marina, Cyprus).
- 1098 ➤ The mean PM₁₀ concentration levels mainly vary from 20 to 50 µg m⁻³ reaching up to 223 µg m⁻³
1099 in Ayia Marina (Cyprus).

1100 In order to describe the vertical structure of the intense Mediterranean dust outbreaks, the CALIOP
1101 vertical profiles of aerosol subtyping and total backscatter coefficient at 532 nm, are used as a
1102 complementary tool to the identified intense DD episodes by the satellite algorithm. Through this
1103 synergistic approach it is found that:

- 1104 ➤ Dust particles are mainly detected between 0.5 and 6 km, following an ascending mode, up to
1105 40° N, leaving from the source areas and transported towards the Mediterranean.
- 1106 ➤ Over the western Mediterranean, the dust layers are mainly observed between 2 and 6 km while
1107 their base height is decreased down to 0.5 km for increasing longitudes.
- 1108 ➤ During the warm period of the year, dust particles are uplifted at higher altitudes (up to 8 km).
- 1109 ➤ In summer, the transported dust loads over the western Mediterranean are recorded above 1 km
1110 and in spring at lower altitudes over the central and eastern parts of the study region. This
1111 behavior underlies the role of topography (e.g. Atlas Mountains) and the enhanced thermal
1112 convection.
- 1113 ➤ The intensity of dust outbreaks, in terms of β_{532nm} , is maximized (up to 0.006 km⁻¹ sr⁻¹) below 2
1114 km and at the southern parts (30° N - 34° N) of the study region.
- 1115 ➤ In spring, considerably high β_{532nm} values (~ 0.004 km⁻¹ sr⁻¹) are observed between 2 and 4 km
1116 in the latitudinal zone extending from 35° N to 42° N.
- 1117 ➤ Moderate-to-high β_{532nm} values are observed up to 6 km, near to the source areas, while the top
1118 of dust layers is gradually decreased down to 4 km towards northern latitudes.
- 1119 ➤ From the longitudinal projection of β_{532nm} , it is evident that DD episodes are more intense (~
1120 0.004 km⁻¹ sr⁻¹) between 1 and 5 km in the western Mediterranean, while over the central and
1121 eastern sectors, the maximum intensities (~ 0.006 km⁻¹ 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_{10} concentrations. For this purpose four intense Mediterranean desert dust outbreaks of different geometrical characteristics that took place across the Mediterranean, namely in Spain (western), Sardinia (central) and Cyprus (eastern), are studied when satellite algorithm's outputs, ground PM_{10} 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_{10} is high. On the contrary, when the dust layer is aloft or its load is not equally distributed in vertical terms then a poor agreement between MODIS- PM_{10} is found.

This study attempts to highlight the importance of the synergistic use of 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.

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 1161 aerosol characterization using available remote sensing datasets.

1162

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1643 **Table 1:** AERONET stations, depicted with cyan colors in Figure 4, which have been used for the identification of desert
1644 dust (DD) episodes based on ground retrievals.

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

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1646 **Table 2:** Percentages of the satellite Ångström exponent, Fine fraction, Effective Radius and Aerosol Index retrievals
1647 satisfying the defined thresholds in the satellite algorithm for the identification of desert dust episodes.

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

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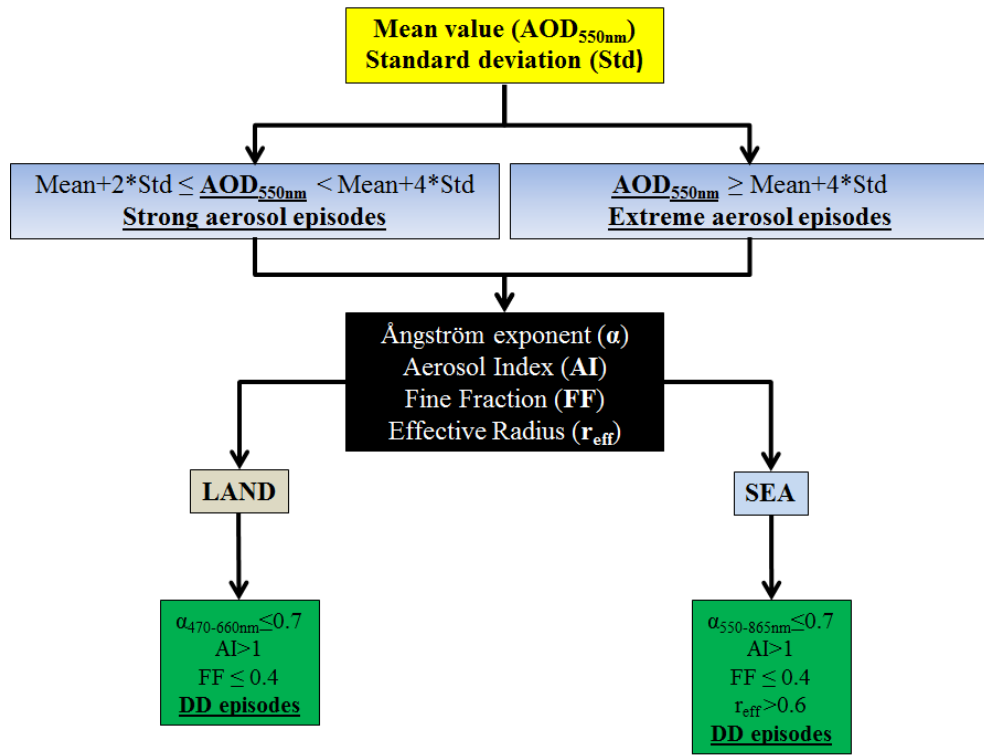
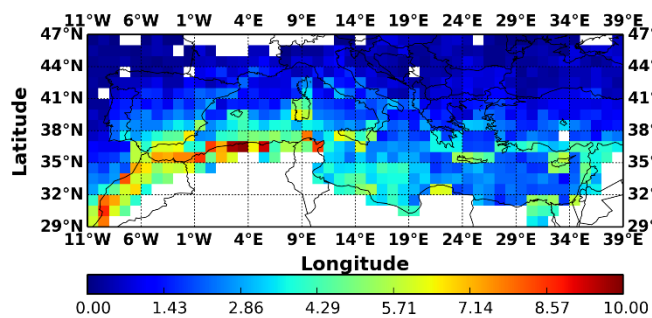
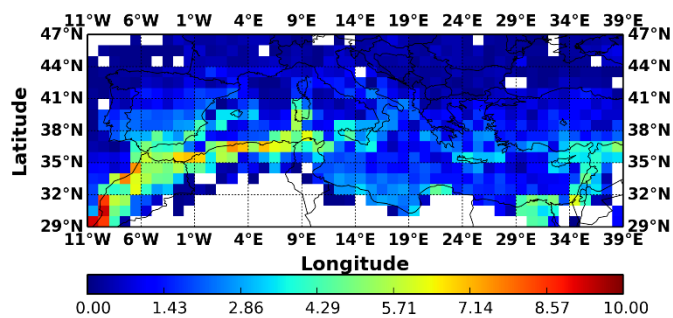


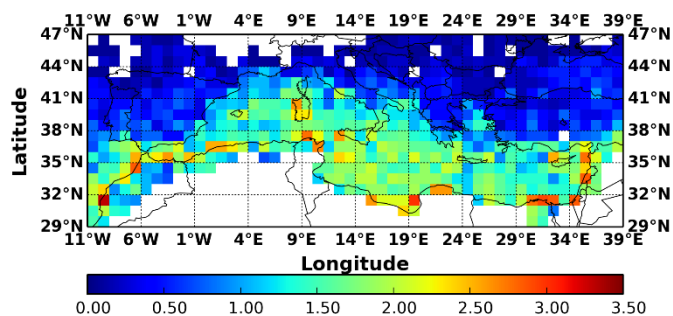
Figure 1: Methodology applied to each 1° x 1° grid cell for the identification of the intense Mediterranean desert dust outbreaks.



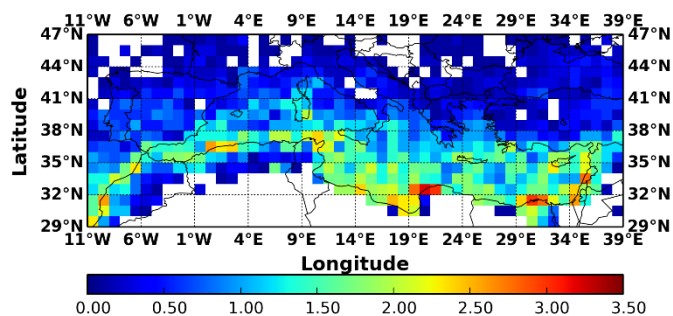
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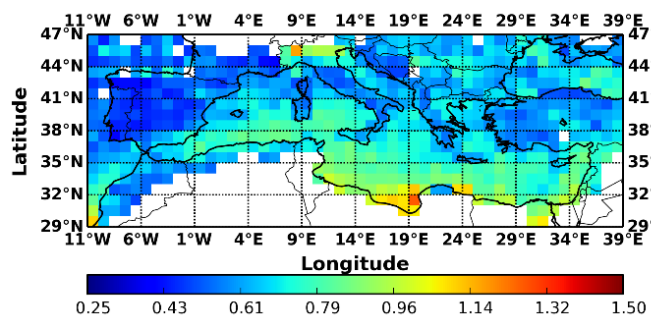


(ii-a)

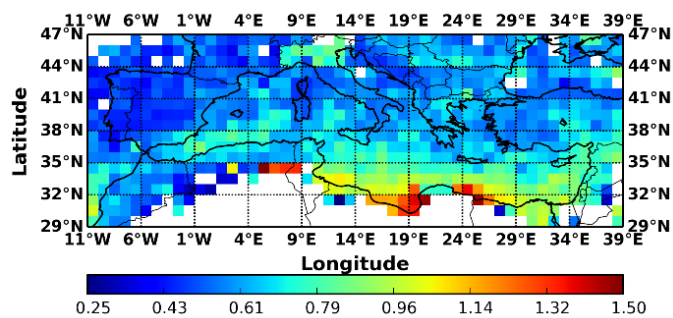


(ii-b)

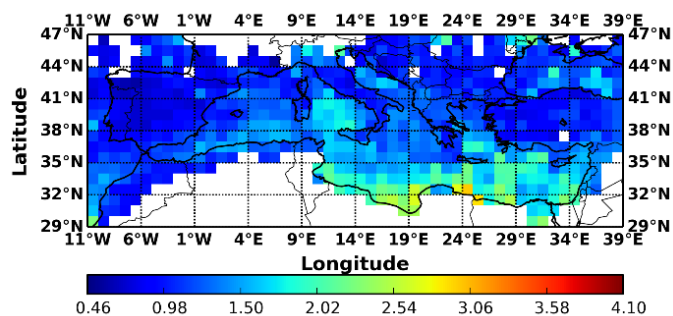
Figure 2: Geographical distributions of the occurrence frequency (episodes/year) of: (i) strong and (ii) extreme desert dust episodes, averaged for the periods: (a) Mar. 2000 – Feb. 2013 (MODIS-Terra) and (b) 2003 – 2012 (MODIS-Aqua), over the broader area of the Mediterranean basin.



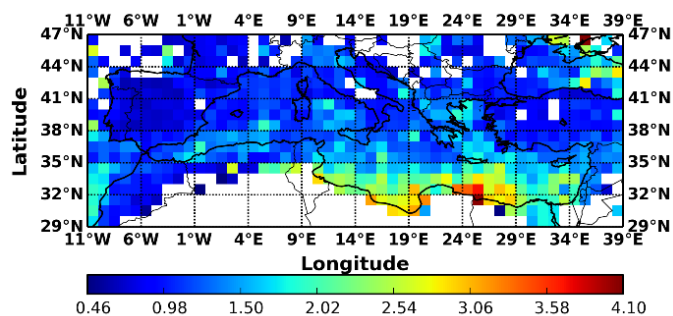
(i-a)



(i-b)



(ii-a)



(ii-b)

Figure 3: Geographical distributions of the intensity (in terms of AOD_{550nm}) of: (i) strong and (ii) extreme desert dust episodes, averaged for the periods: (a) 2000 – 2013 and (b) 2003 – 2012, over the broader area of the Mediterranean basin.

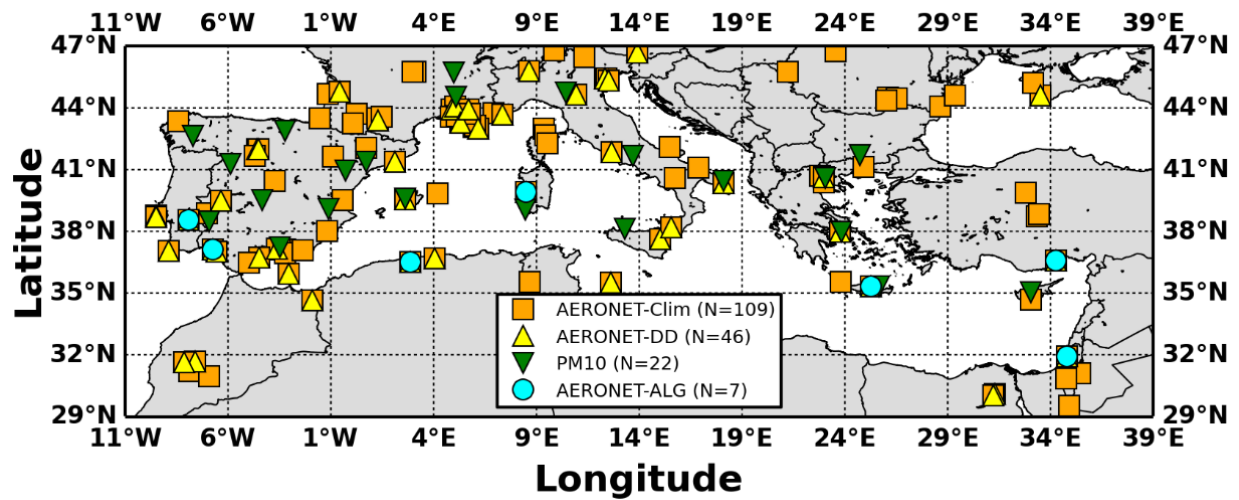
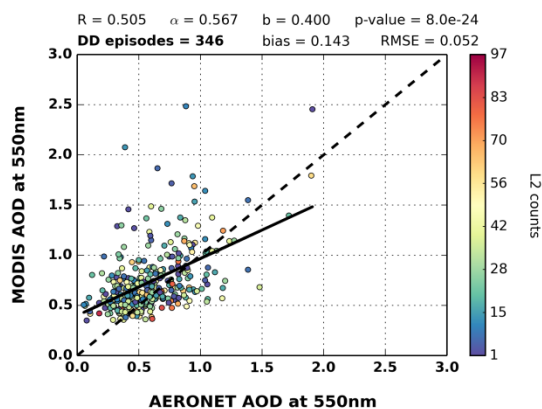
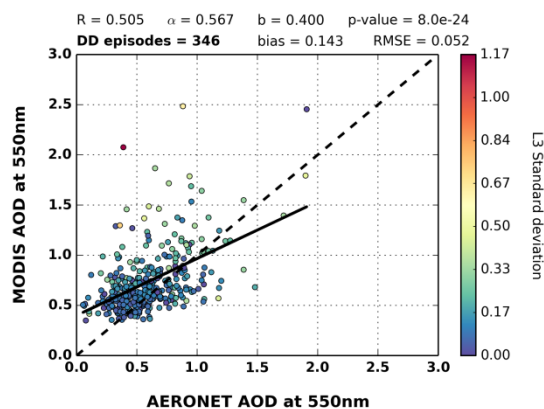


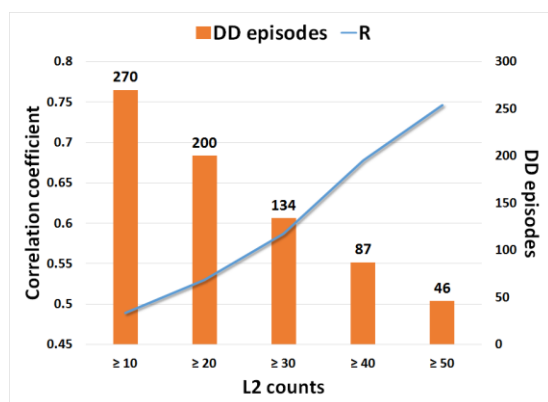
Figure 4: Locations of the AERONET and PM₁₀ stations which have been used for the evaluation of the algorithm's outputs. More specifically, with orange squares are denoted the AERONET stations located into the study region, with the yellow triangles the AERONET stations with coincident satellite and 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 PM₁₀ stations.



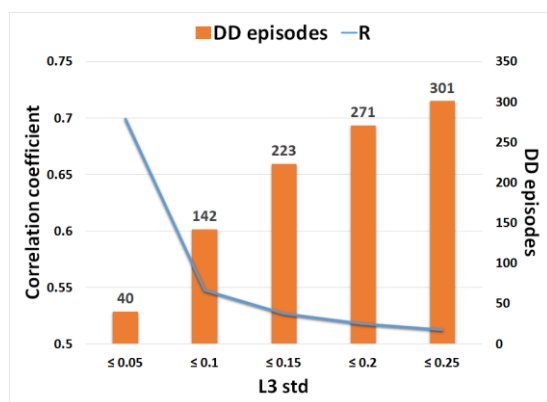
(i-a)



(i-b)



(ii-a)



(ii-b)

Figure 5: (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.

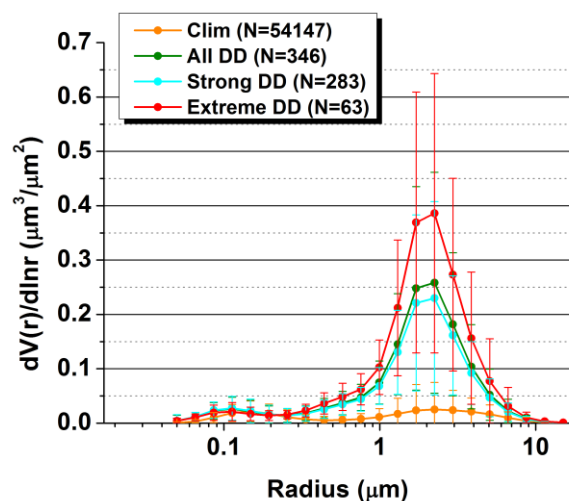


Figure 6: AERONET size distributions averaged for all available retrievals (orange curve) as well as for the total (green curve), strong (cyan curve) and extreme (red curve) desert dust episodes, occurred over the broader area of the Mediterranean basin, during the period Mar. 2000 – Feb. 2013. The error bars represent the calculated standard deviation.

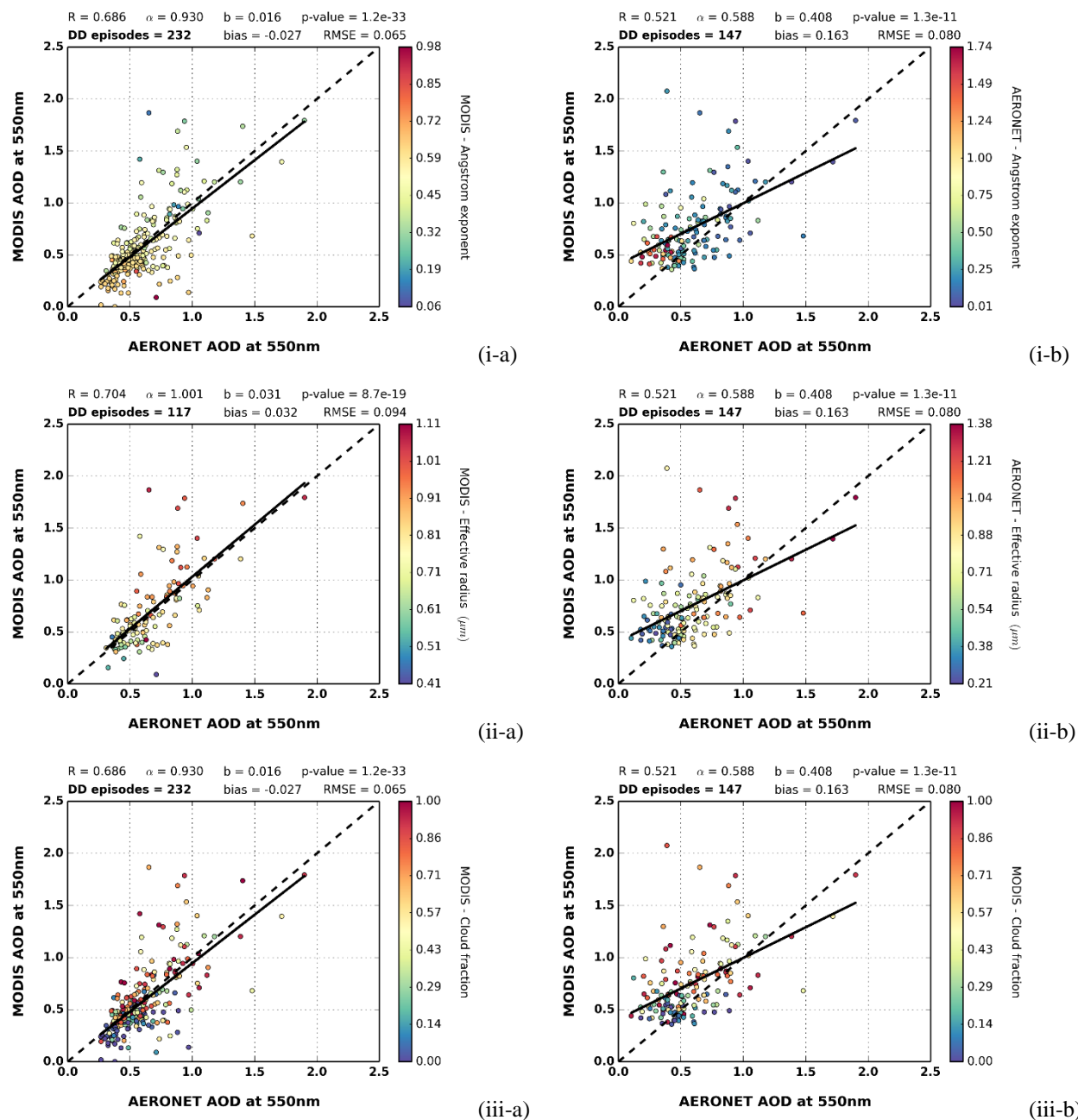


Figure 7: Scatterplots of MODIS-Terra and AERONET aerosol optical depths at 550 nm when intense dust episodes have been identified based on: (a) AERONET retrievals and (b) satellite algorithm, respectively. In the left column, colormaps indicate the corresponding values of: (i) Ångström exponent, (ii) 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 (α), intercept (b), p-value, number of DD episodes, bias and root mean square error ($RMSE$).

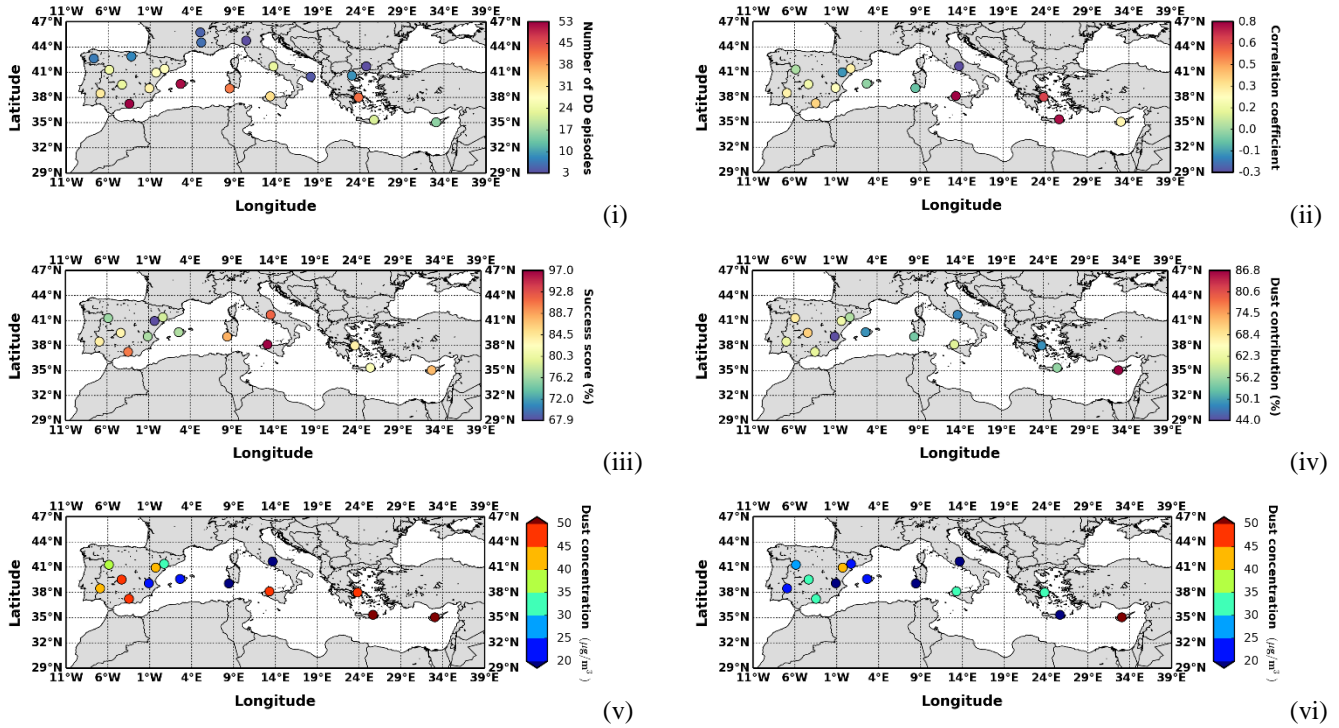


Figure 8: (i) Number of concurrent intense DD episodes where total PM_{10} concentrations and MODIS-Terra AOD retrievals are available, (ii) Computed correlation coefficient values between total PM_{10} concentrations and MODIS-Terra AOD retrievals in stations where at least 10 DD episodes have been 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.

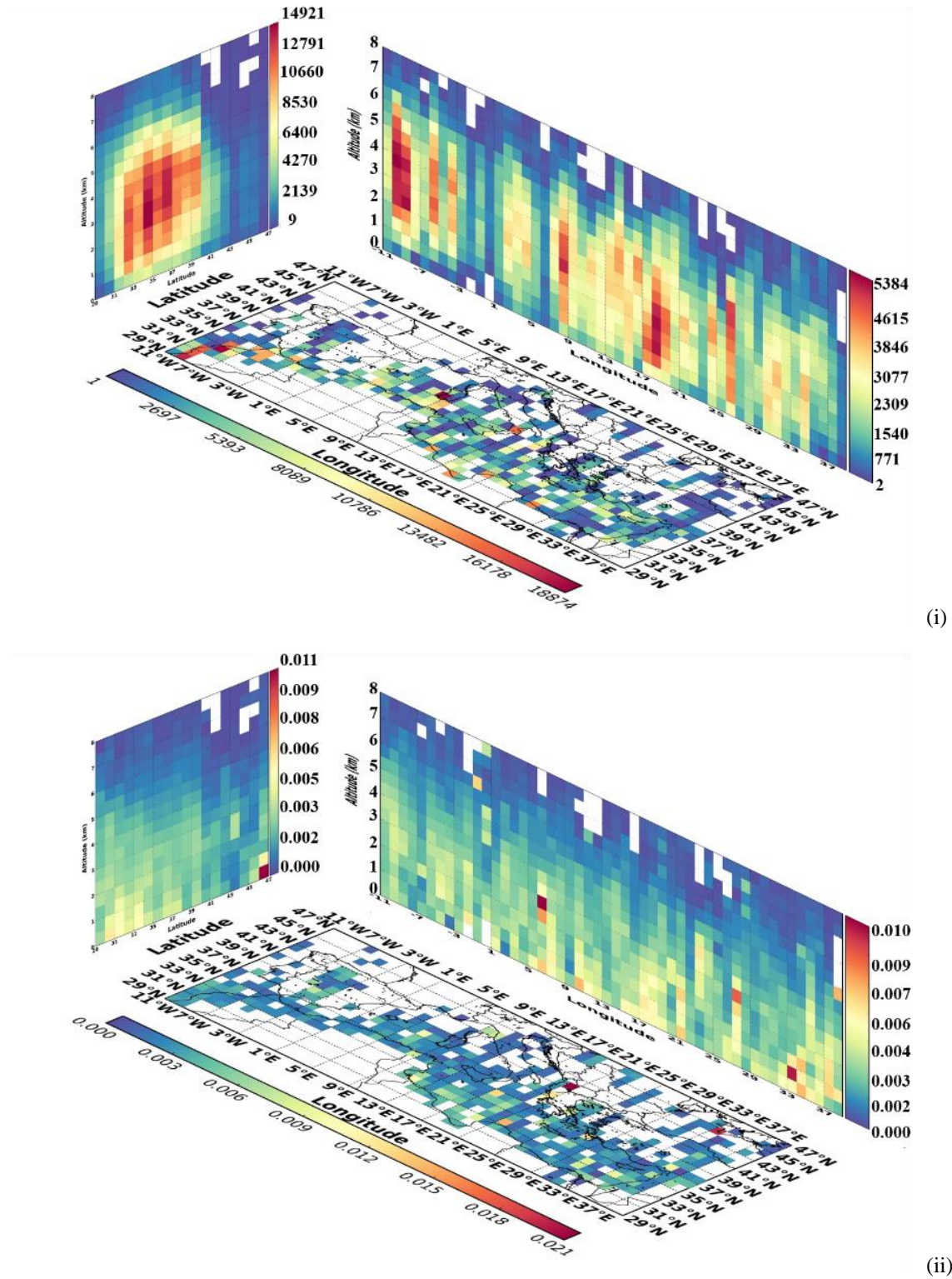
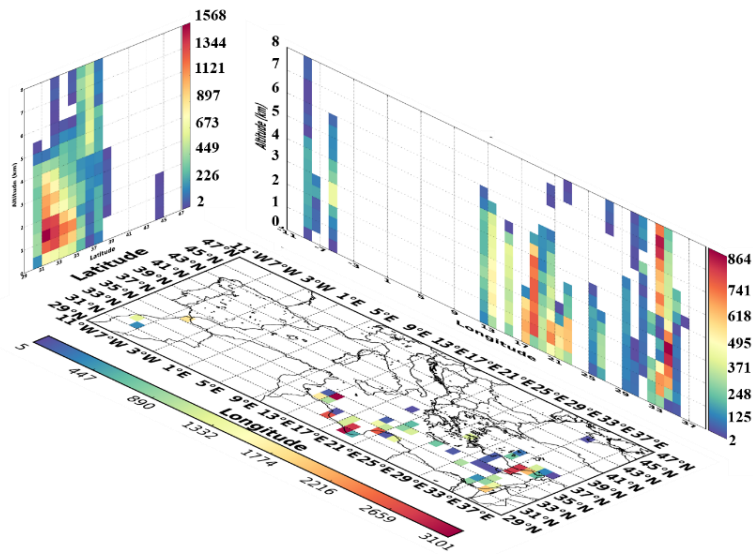
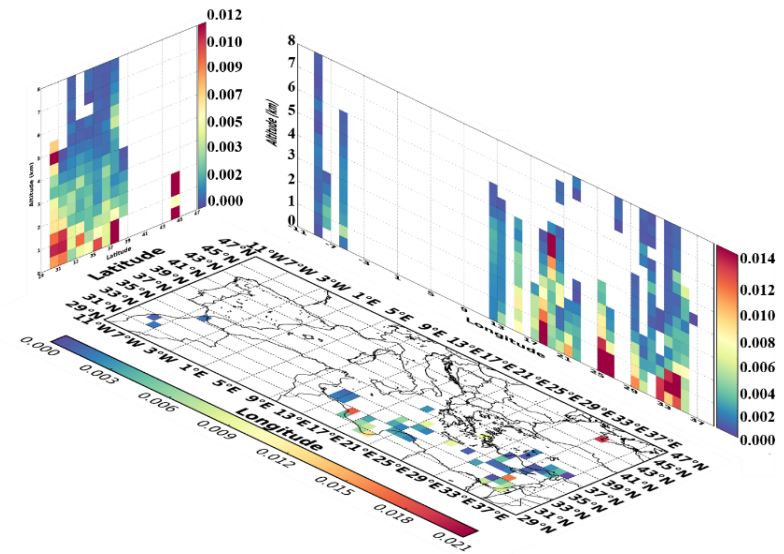


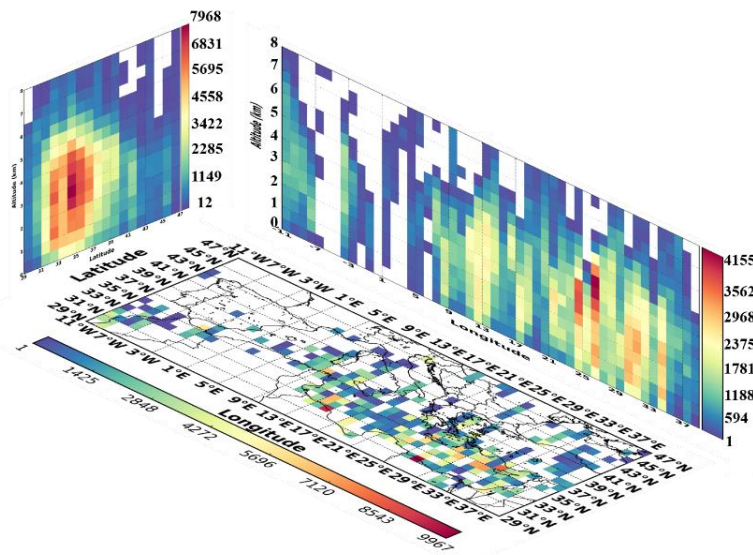
Figure 9: Three dimensional structure of the: (i) overall number of dust and polluted dust observations and (ii) total backscatter coefficient at 532 nm ($\text{km}^{-1} \text{sr}^{-1}$), over the broader Mediterranean basin under DD episodes conditions, based on CALIOP-CALIPSO vertical resolved retrievals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



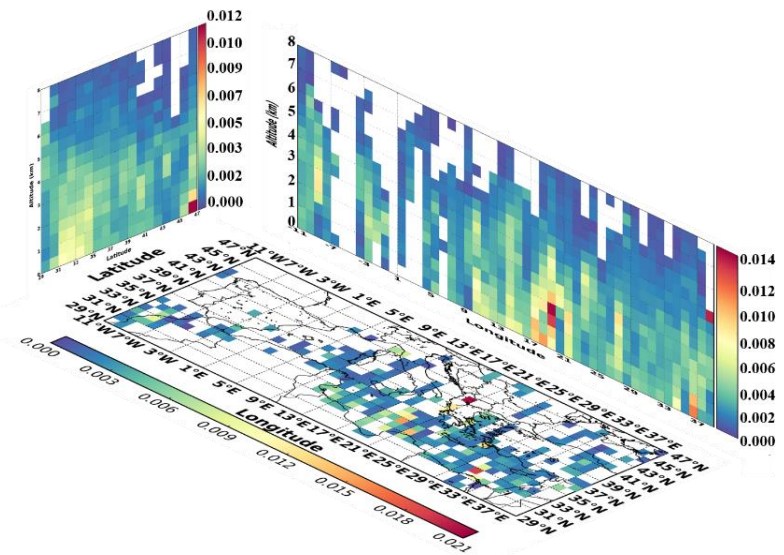
(i-a)



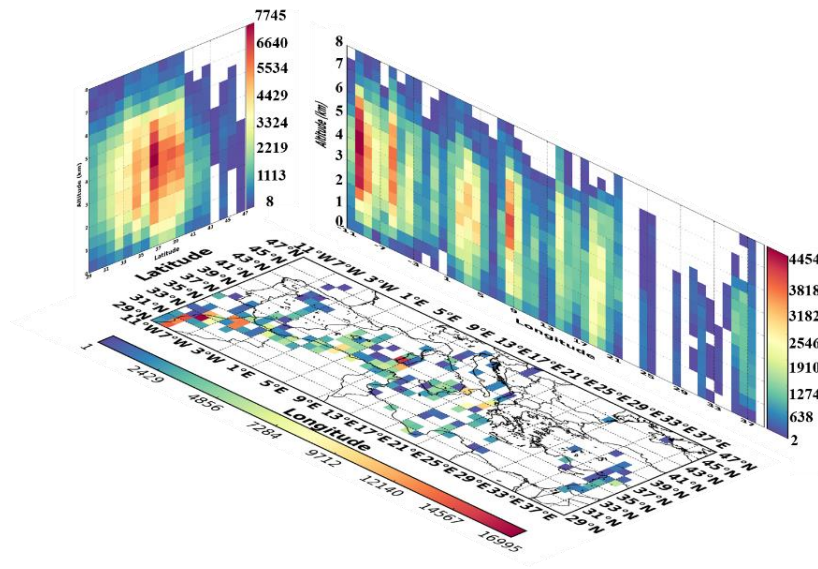
(i-b)



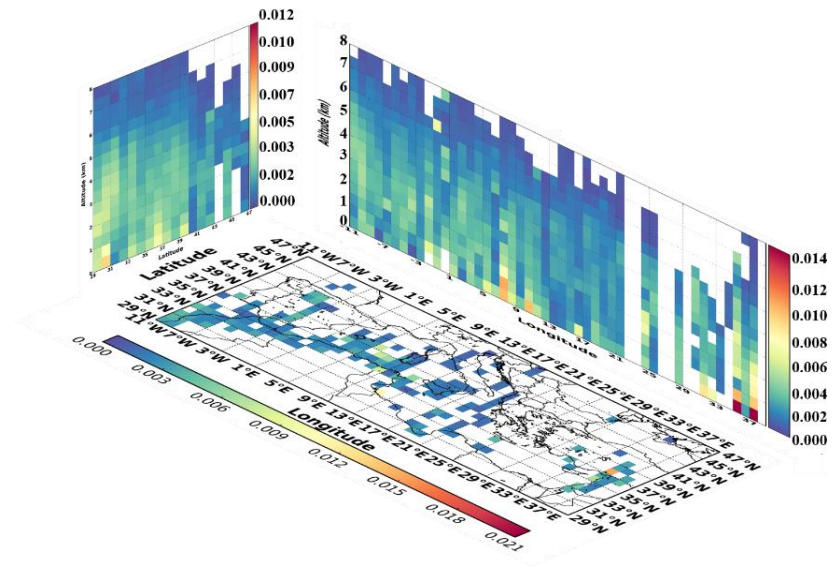
(ii-a)



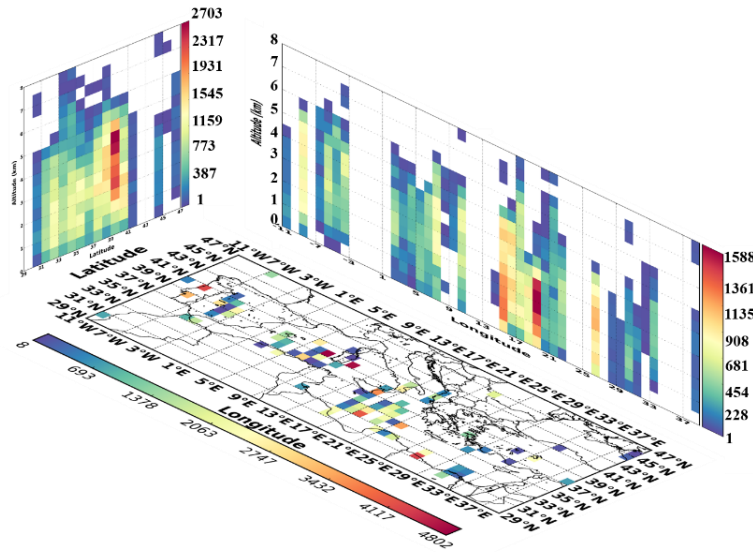
(ii-b)



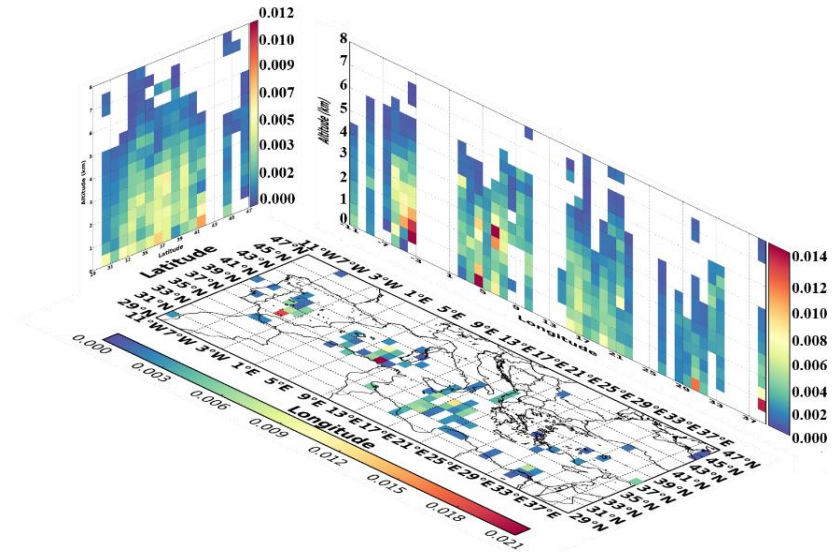
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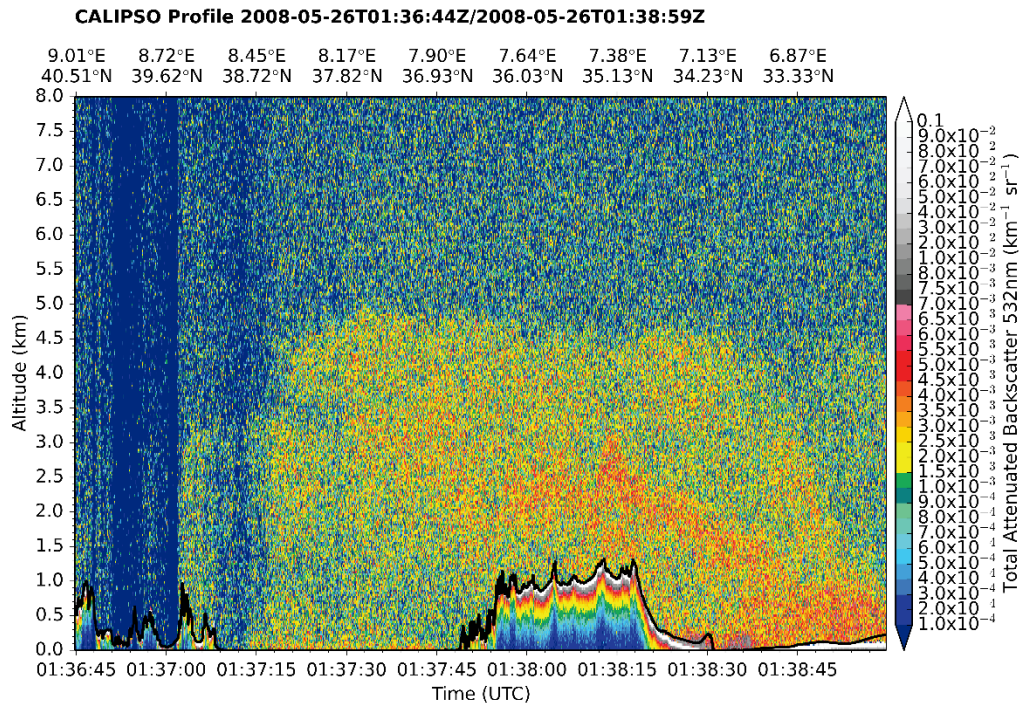


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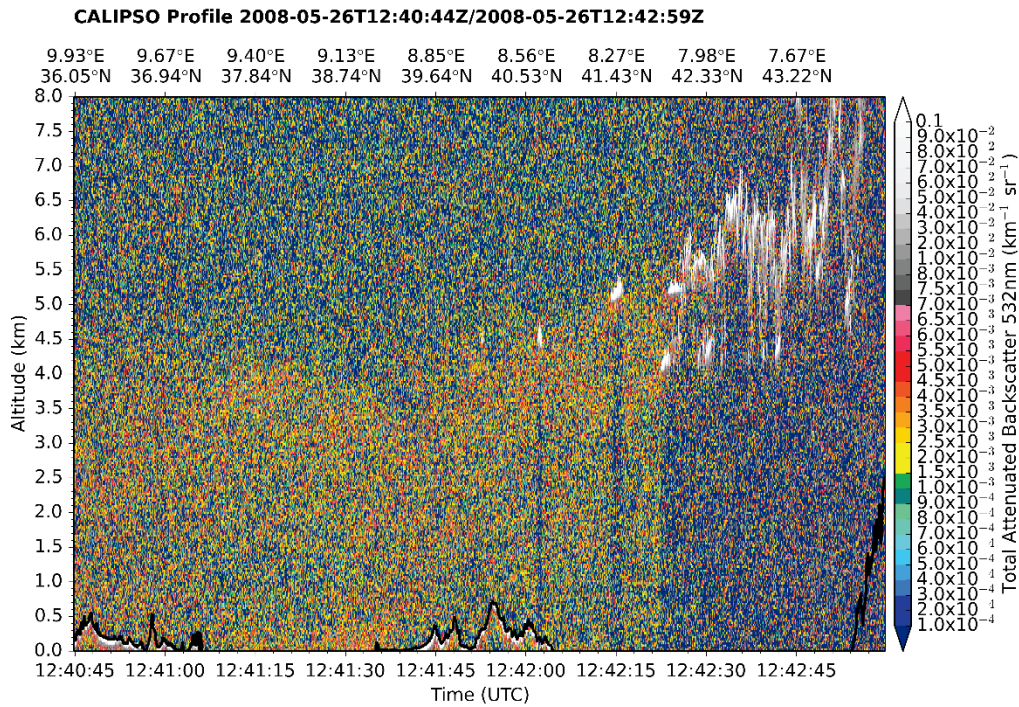


(iv-b)

Figure 10: 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 vertical resolved retrievals, over the period Jun. 2006 – Feb. 2013.

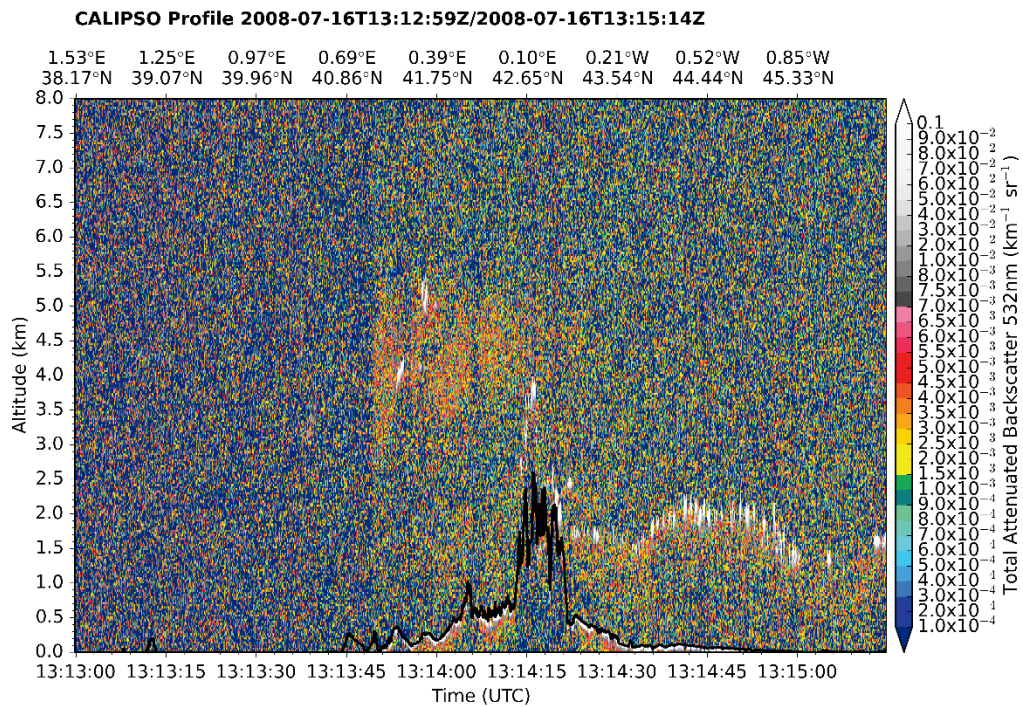


(i)

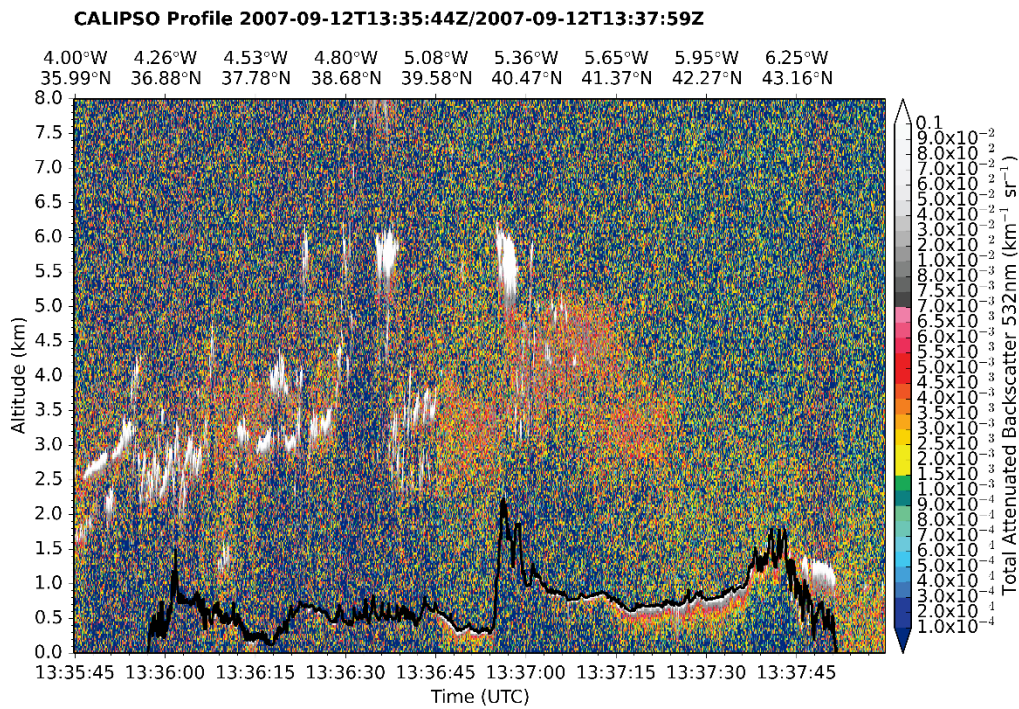


(ii)

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: (i) nighttime and (ii) daytime, on 26th May 2008, over the station Censt (Lat: 39.064, Lon: 8.457). The black thick solid line represents the surface elevation.

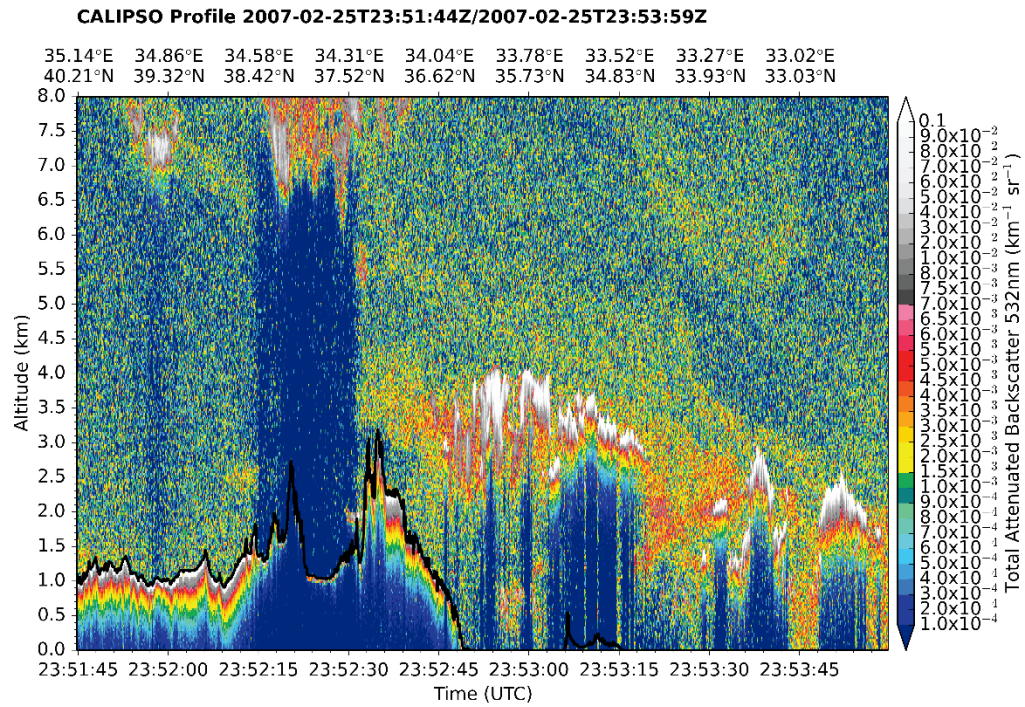


(i)



(ii)

Figure 12: 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 16th July 2008 and (ii) San Pablo (Lat: 39.525, Lon: -4.353) on 12th September 2007. The black thick solid line represents the surface elevation.



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1801 **Figure 13:** Cross section of the total backscatter coefficient at 532 nm (in $\text{km}^{-1} \text{sr}^{-1}$) vertical profiles along the CALIOP-
 1802 CALIPSO track during daytime over the station Agia Marina (Lat: 35.039, Lon: 33.058) on 25th February 2007. The black
 1803 thick solid line represents the surface elevation.

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