Response to Editor

We would like to thank the Editor for his comments helping us to improve furthermore our paper. Below, you can find our comment-by-comment replies.

"*Thank you for your efforts in improving the initial version of your manuscript. I recommend that you submit a revised version considering the new comments made by referee #2 (see initial report #4), and the additional comments in my initial report that were not yet considered.*"

We would like to clarify that many comments (e.g. statistical significance, potential reasons of DD episodes' misclassification) from the Editor's initial report were already included in the submitted document. Here, in the revised manuscript, we have added a short text (Lines 499-505) referring to the comparison of the old (Gkikas et al., 2013) and new version (present analysis) satellite algorithm outputs versus AERONET retrievals, as it has been suggested by the Editor. This was done in order to investigate the benefits of using QA-weighted level-3 MODIS AOD data (present analysis) in contrast to non-weighted QA ones (Gkikas et al., 2013). Note that the utilization of QA-weighted MODIS retrievals constitutes an improvement of our algorithm and for this reason any possible impacts on its performance must be evaluated.

"I share with this referee the feeling that a number of clarifications given in your reply letter in response to our comments on the initial version would be useful to the readers and need to appear in the paper itself. One example concerns your reply on the use of the cloud fraction (CF) test. The main text (line 342) still presents the use of this test as an improvement of the methodology, when your reply indicates that this filter is only performed for a sensitivity test. Please clarify."

The majority of the comments made by the Reviewer-4 were primarily related to the applied methodology (satellite algorithm) and secondarily to data description and are more linked to our published paper in ACP (Gkikas et al., 2013). In the submitted manuscript, despite the disagreement of many of the authors to accept all the argumentation without any cited work to back it up, we have included as many as possible of our replies, addressing issues raised by the Reviewer-4, to the text resulting thus in an extension of its length. Moreover, the extension of the manuscript parts related to the satellite algorithm and data (both discussed thoroughly in our previous papers) induces an unbalance and inhomogeneity making hard to a potential reader to focus on our primary scientific target, which is the description of dust outbreaks' vertical structure. This is clearly reflected by the fact that half of the Results section is dedicated on the relevant discussion (Sections 4.3 and 4.4). For this reason, we have selected to insert to the document the most important replies, to our opinion, and not all of them. As it concerns the second part of the Editor's comment, we agree with him and we have modified the relevant part of the manuscript accordingly (Lines: 419-424).

"I also believe an effort must be made on the structuration of the paper which is long, trying to decrease the number of sub-titles levels. I invite you to reconsider the referee and my own initial comments on the structure of the paper. For instance, I recommend that the methodological points that I listed in my initial report are not left in the results section. For instance what concerns AERONET

can come in the description of AERONET data (e.g., derivation of AERONET AOD at 550nm) or in a small additional paragraph at the end of section 3 (e.g., the detection of dust episodes with AERONET data). I also follow the referee suggestion to present first in section 4 the comparisons/"evaluation" of the algorithm before going to main results on the horizontal and vertical distribution of dust episodes. For this, I think that the section 4.2 would better come as a sub-section of present section 3 or make a distinct section (4) before the Results section (5) itself. Furthermore, I suggest to have two distinct main sections discussing results, a first one (5) made of the geographical distributions (present section 4.1) with 2 sub-sections on frequencies and intensities, and a second one (6) on the vertical distribution including present sub-sections 4.3 and 4.4, the last one being presented as a series of case studies without reference to PM in its title."

We agree with the Editor that the length of the manuscript is long but consider that, to our opinion, we are providing the information which is required in order to support our findings helping thus the reader to follow our approach. It is evident, that the length of the text has been grown due to the inclusion of our responses to the Reviewer-4 comments as well as to the added Section 4.4 which has been suggested by the Reviewers 2 and 3. In addition, during the second round of comments, new questions have been raised by the Reviewer-4 and the inclusion of our responses to the revised document will cause a further extension of its length. Trying to compensate this, we had removed from the initial document submitted to the ACPD, three figures related to AERONET (now available in the supplementary material, Figs. S2-S4) and the obtained results are briefly (less than one page) discussed in Section 4.1.1.3. Also, we would like to point out that through the review phases (your initial report, the four reviews, your second report and the second round of reviews) we have modified the text accordingly trying to address all the reviewers' proposals which in some cases were contradictory. The most outstanding example is that the Section 4.4, suggested by the Reviewers 2 and 3, should be removed according to the opinion of the Reviewer-4.

As it concerns the structure of the paper, in the revised manuscript we have changed the order of Sections 4.1 and 4.2 as it has been proposed by the Editor. More specifically, the comparison of the satellite algorithm's outputs against AERONET/PM_{10} comes first, then the geographical distributions of DD episodes' frequency/intensity are drawn while in Sections 4.3 and 4.4 are discussed the results for the vertical structure and the level of agreement between MODIS-PM, respectively. Nevertheless, we would like to clarify the reasons why we have not fully adopted the Editor's suggestions. We believe that is better to keep Section 3 as is since it is described the identification of the desert dust episodes based on the satellite algorithm, which is the main tool used in the present analysis. In addition, we believe that the evaluation of the satellite algorithm must be a sub-section in Results and not in a separate section since useful information, based on AERONET retrievals (e.g. volume size distribution), is provided. Moreover, we think that it is not necessary to split the section where the geographical distributions of the DD episodes' frequency and intensity are presented in two subsections in order to avoid adding new subtitles levels. As it concerns the "methodological" parts of the manuscript, we strongly believe that the existing serial order which we have decided helps the reader to understand our approach in each phase of the analysis. We are aware that this issue was raised in your initial review but still we prefer to sustain the existing structure of the paper. We believe that is quite important considering that a potential reader will not be forced to go back and forth throughout the manuscript. For this reason, in Section 4.1.1.1 we are discussing briefly how the AERONET AOD_{870} retrievals are converted to AOD_{550}, through the implementation of the Ångström formula, in order to match with the corresponding MODIS data. For the same reasons, in Section 4.1.1.4 (identification of DD episodes based on AERONET), Section 4.3 (satellite algorithm – CALIOP) and Section 4.4 (specific dust cases) the reader can find all the relevant information gathered (followed methodology – obtained results). Summarizing, we think that it is a matter of personal point of view and we would like to keep ours. Our argument is also supported by the fact that the Reviewers 1-2-3 agree that the presentation of our results is clear and the paper is well structured. Finally, we have removed from the titles the PM sites' names as it has been asked by the Editor.

"I also follow referee 2 on the fact that the PM stuff has no major input for the main purpose of this study and could be removed for shortening and better focusing the revised paper."

We disagree with this comment for the following reasons. The part of the manuscript referring to the comparison between satellite algorithm's outputs and PM_{10} concentrations occupies less than 1.5 pages. This means that its omission will not reduce significantly the length of the manuscript. On the contrary, apart from the results which are presented here (success scores, dust contribution, mean and median levels) and not in Gkikas et al. (2013), thanks to this analysis we are investigating the level of agreement between columnar (MODIS) and surface-based (PM_{10}) measurements across the Mediterranean. In a further step, the obtained results trigger the analysis which is presented in Section 4.4 where it is investigated (through the CALIOP-CALIPSO lidar retrievals) how the desert dust outbreaks' vertical distribution affects the level of agreement between columnar (MODIS AOD) and ground (PM_{10}) observations. For the aforementioned reasons, we believe that the omission of Section 4.1.2 will cause a discontinuity, not helping us to clarify to a potential reader why/how we have selected the specific desert dust episodes which are analyzed in Section 4.4. Moreover, considering the extension of Section 3, by adding to the revised manuscript our old/new responses to the Reviewer 4, the removal of Section 4.1.2 will induce an unbalance (in terms of text length and focus) between the obtained results and the discussion about the applied methodology. In addition, if we follow the Editor's suggestion then the additional information of the present analysis with respect to Gkikas et al. (2013) will be minor to our opinion and we would like to avoid it. Of course, we can remove Section 4.1.2 from the revised manuscript just citing the obtained results from Gkikas et al. (2013) but in this case someone easily can raise a question if the aforementioned findings are similar for the extended period (almost double years) considered in the present analysis.

"Results on the horizontal distribution need to appear in the abstract."

The results about DD episodes' frequency of occurrence and intensity were already appeared in the abstract. Please, see lines 32-41.

Other technical remarks:

"-Fig.1 is almost identical to Fig. 2 published in the same journal by Gkikas et al. (2013). Please specify "(after Gkikas et al., 2013)" in the figure legend."

We have decided to remove Figure 1 from the manuscript following your initial proposal in the uploaded comment to the ACPD (1st March). In the revised manuscript, we are directing the reader to Figure 2 (in Gkikas et al., 2013) where the flowchart of our methodology is depicted.

"-Please check the systematic use of the italic style for abbreviations, especially SSA and AOD."

We have changed to italic format all the SSA and AOD abbreviations.

"-add missing doi numbers in the reference list (especially verify J. Geophys. Res. papers)."

We have added the doi numbers which were missing from the reference list.

"-Abbreviate "Remote Sens. Lett." in line 1189, "Ocean." in line 1347, "Technol." in lines 1400, 1463 and 1609, "Trans. Inst. Br. Geogr." in lines 1433-1434, and "Adv. Meteorol." in lines 1442-1443."

Done.

"-Also check missing or incorrect journal name in the references Matthias et al., 2004 ("J. Geophys. Res."), and Papayannis et al., 2014 ("Sci. Total Environ., 500-501,"...); check also the spelling of the 2nd author "Nicolae" in Papayannis et al., 2014)."

Done.

"-no italic in line 1521."

Done.

Response to Reviewer-4

We would like to thank the Reviewer for his/her second round of comments, which we tried to take into account in order to further revise and improve our manuscript. Our responses (in regular format) to the comments (in italic format) are listed below.

General

"Many answers to my questions are given as 'reply to the reviewer', but not (or only partly) inserted into the body of the manuscript itself. It was obviously the intent of my revision to provide hints to strengthen the manuscript, and make it more legible. I did not need 'personal' answers, my comments were rather oriented to make the authors provide any potential reader with all the needed/useful information and to show/demonstrate the robustness of the methodology proposed to achieve the results. Thus, some of my comments were not fully understood/addressed in the text and this intent of mine was therefore not achieved within the revised version submitted. My personal impression in reading the revised version is that all the efforts done by the authors in revising the text translated at last into some additional material inserted within the original manuscript in a rather 'patchy' way, with no substantial improvement of its structure and no real embedding of the given suggestions in its core concept. The overall result is a long text, focusing on (too) many different aspects. The authors should make an effort to decide which is the core objective of the manuscript and limit the length of the other sections. Some suggestions on which parts could be removed (and possibly addressed in a companion study) were already given in my first review, and are further indicated below."

First of all, we would like to kindly remind that this Reviewer is and has been not solely reviewing our manuscript, and that there are and have been three others. Thus, our revision was based on the comments of all of them, which not always were in line with this Reviewer comments. This could at least partly respond the Reviewer's statements referring to the improvement and re-structure of the paper according to his/her indications. In addition, we would also like to kindly remark that our responses in the previous round and the associated modified manuscript were not supposed to just show that every comment of this Reviewer has been adopted and inserted in the manuscript; it is our opinion that this is an important principle in the review/revision procedure of a paper. Of course, we believe that whenever responding to a Reviewer comments, authors must show that all of them are considered, not just adopted, while efforts should be made to either implement them in the revised manuscript when adopted or appropriate replies and arguments are given in the answer when not endorsed.

In this line, we believe that we have made a strong effort to give detailed answers to all of the Reviewer's questions and to insert as many as possible of them to the revised manuscript. However, we also took care not to extend the paper length with information that is either already provided in our previous papers or it is not critical, to our opinion, for a potential reader. We would like to remind the employed methodology in this paper does not appear for the first time but has been introduced to a few previous evaluated and published papers in other ISI journals or to ACP.

The majority of the Reviewer's comments in his/her first report were mainly focused on the applied methodology (satellite algorithm) and to the data description. Trying to address and implement the Reviewer's old and current raised issues we were and still are forced to extend the aforementioned parts of the manuscript, sometimes risking to make it probably unbalanced and inhomogeneous. If we are to follow the Reviewer's suggestion to remove from the paper the PM analysis as well as Section 4.4 (which has been suggested by the Reviewers 2 and 3) then indeed the length of the paper would be shortened, but on the other hand this may result in a much similar paper with that by Gkikas et al., (2013) and focused on discussing issues related to the applied methodology. This was/is not the main objective of this paper, in which we really intend to improve our methodology but also emphasize the vertical structure of the Mediterranean desert dust outbreaks. According to this strategy, first an analysis is undertaken to ensure that the satellite algorithm's outputs are reliable, also highlighting the possible differences between the current geographical distributions and the corresponding ones obtained by Gkikas et al. (2013). Next, the CALIOP vertical resolved dust retrievals are used to investigate the Mediterranean dust outbreaks' annual and seasonal characteristics (in Section 4.3) as well as to give a better insight of how the vertical distribution of dust loads affects the level of agreement between columnar (MODIS) and ground (PM_{10}) measurements (in Section 4.4). We believe that the present manuscript is organized in the best possible way to fulfill the afore-mentioned priorities in this paper.

More specific answers to the Reviewer's comments referring to methodological and datasets problems, as well as to the presentation of results are given below. As it has been mentioned above, we have decided to sustain the PM_{10} analysis as well as Section 4.4 in the revised document in contrast to the Reviewer's suggestion for the reasons which will be discussed later.

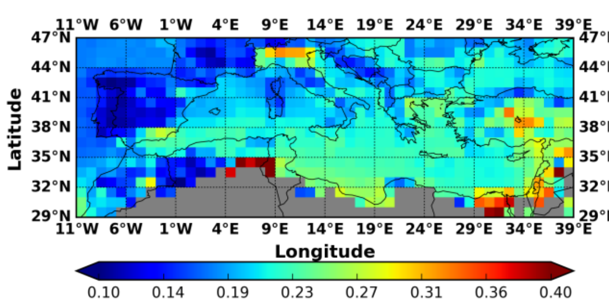
1. METHODOLOGICAL PROBLEMS

"An unequal distribution of AOD data within the whole dataset considered DOES HAVE A CRITICAL IMPACT on the pixel-resolved 'AOD Mean values' being derived and THEREFORE on the final outcome of their algorithm. Proving there is a rather uniform distribution of the number of single, daily-resolved AOD data points in the dataset is essential to demonstrate your method builds on robust basis. Should this not be the case, i.e., should the number of AOD data-points be unbalanced (in the different months or in the different years within the long-term period covered), other strategies should be adopted to compute reasonable 'AOD Mean values' (i.e., the threshold values used to drive the empirical algorithm)."

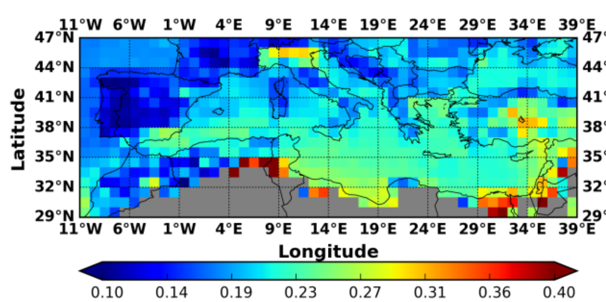
The applied methodology which is followed in the present analysis for the calculation of the mean AOD levels has been used in numerous studies in the past dealing with satellite or AERONET data at local, regional or global scale. It is well known that the satellite and ground aerosol retrievals are characterized by an unequal distribution of available data (in spatial and temporal terms). We agree with the Reviewer that this temporal inhomogeneity can affect the calculated threshold levels but their definition as well as the identification of DD episodes is simply done based on the available data. However, following the Reviewer's thinking then we have to reject thousands of papers dealing with AOD climatologies/trends studies just because satellite (e.g. MODIS) or ground (e.g. AERONET) data are utilized which are unequal distributed on monthly, seasonal, annual or other possible temporal scale.

This “loss” of data would lead to lower mean AOD levels (based on our default methodology) if for example a dust outbreak had occurred during the missing day/s. In addition, the calculated mean AOD levels will be positive or negative biased depending on the seasonal availability of AOD retrievals. We can understand the Reviewer’s argument if the obtained results are going to be used for health impact studies but we disagree as it concerns the climatic effects. In his/her initial report he/she suggested to compute the mean AOD levels from monthly values (weighted equally) instead of daily ones as we are doing in our methodology. In case of radiative forcing studies (related to climatic impacts) the direct radiative effects on a monthly basis, in all studies to our knowledge, are calculated according to the available daily measurements of each month. Following the Reviewer’s suggestion, the normalization of AOD requires at the same time the normalization of the identified dust events on a monthly scale which is not feasible due to the variation of AOD retrievals’ availability (attributed to clouds, retrieval algorithm assumptions) as well as by the seasonal variation of dust events. Moreover, it is not clear why the mean AOD levels should be calculated by monthly values and not from seasonal or annual ones or other temporal scales (e.g. weekly).

However, in order to be more specific and dispel any concern, we proceeded as suggested by the Reviewer, namely we calculated the mean AOD levels (on which are based the AOD thresholds) based on monthly values (calculated by any available daily retrievals) and we have compared them against the corresponding ones calculated based on our default methodology (daily retrievals). According to our results, depicted in Figure R1, the two sets of AOD mean values have almost identical spatial patterns and magnitudes. The differences are smaller than 0.1 in absolute or 5% in relative terms. More specifically, the monthly-based mean AODs over land are smaller than the daily-based ones by less than 10 % (observed in northern parts of the study region where dust outbreaks have less impact) while the opposite is found over sea. This finding reveals thus that the unequal temporal distribution of daily AOD retrievals does not have a critical impact on the computed mean AODs (Lines: 372-381) utilized in this manuscript. Therefore, also taking into account the common practice and for reasons of consistency with our and others’ previous studies we prefer to keep using the daily-based AOD mean and threshold levels.



(i)



(ii)

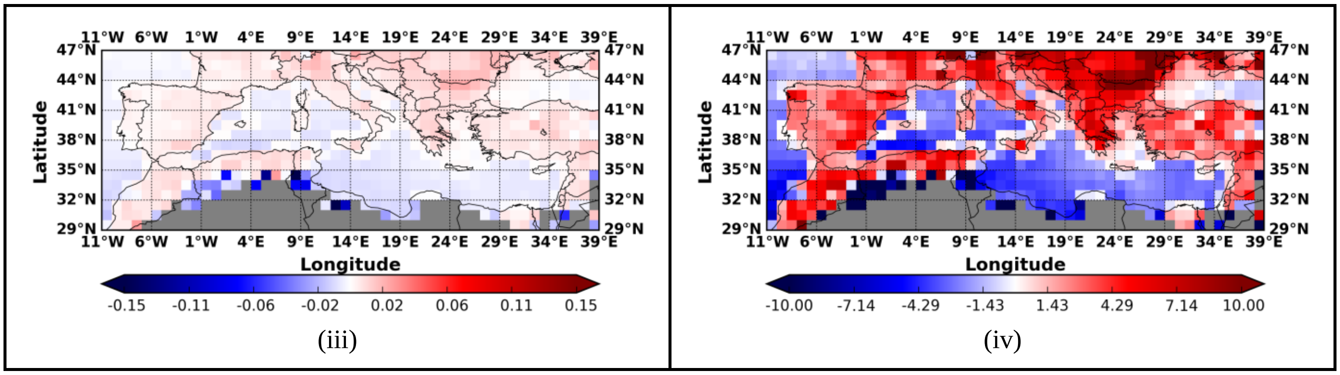


Figure R1: Geographical distributions of the: (i) long-term averaged AOD_{550nm} based on daily MODIS-Terra, (ii) long-term averaged AOD_{550nm} based on monthly MODIS-Terra, (iii) absolute daily-monthly differences and (iv) percentage daily-monthly differences. The calculations for the broader Mediterranean area have been made over the period 1 Mar. 2000 - 28 Feb. 2013.

Summarizing, this point, looking at any published AOD map of any satellite product, someone can argue that the map has no meaning since it is produced by an unequal distribution of AOD data over different days/months/years for different grids. To take this even further, even the 1 by 1 degree daily MODIS AOD could be considered in general, inhomogeneous since it is the average of 10 by 10 Km AODs that represent different areas during different days due to the day to day random presence of clouds in the 1 by 1 degree pixel. Does this mean that any AOD time series by any satellite in any 1 by 1 grid has no scientific importance before a spatial homogenization/normalization?

Continuing the normalization discussion for CALIPSO data, things get much worse as a CALIPSO daily/monthly profile in any publication is a sum of a number of parts of vertical profiles in a specific grid. In this case in addition to the temporal and the 2D spatial inhomogeneity, a 3D inhomogeneity is included.

Our definition of the AOD mean calculated with only available data is based on answering the main question of the work which is if at a certain day and a certain grid we have a strong or an extreme dust event.

"In fact, in Figure R1-to-R4 the authors show the data in terms of 'percentages', without reporting the formula these percentages represent. Anyway, if I understand correctly, this 'percentage' means that for example in the month of January (Figure R1) a 100% value (dark red) is obtained in pixels when they get 31/31 daily data points for each year of the period considered (i.e., they get (31 days x 13 years)/(31 days x 13 years) values, covering all days of January 2001-2002-2003-2004-2005-2006-2007-2008-2009-2010-2011-2012-2013). Is this interpretation correct? If no, the authors should please define what's the meaning of 'percentage' here. If yes, then by similarity your Figure R2 shows that in winter they get more than 85% of the data over sea and less than about 50% over land (excluding bright areas having no data), while the opposite occurs in summer. This clearly reveals the seasonal unbalance I suspected, plus a land/sea unbalance in data availability I did not expect. This does critically affect their results."

Yes. The calculated percentages refer to the availability of the daily MODIS-Terra AOD retrievals depending on the temporal scale (i.e., months, seasons, years and whole study period). As shown in the previous response, the seasonal unbalance does not critically affect our results.

"Why should cloud cover in winter affect land and not sea (Mediterranean) areas? Are the two different MODIS retrievals over land and ocean playing a role in this land/ocean difference in the number of data availability? It is true that the DD impact in winter at higher latitudes is expected to be low, but what about the different data availability over the Med sea among seasons? This clearly affects the results over the Med areas closest to the North African coasts where the maximum impact of dust is rather expected. The authors should comment on that."

The data availability varies between land and sea surfaces due to the different applied MODIS retrieval algorithms. During warm seasons, across the Mediterranean Sea, a large amount of retrievals is masked out from the dataset due to Sun glint, which varies with season and gets less important in winter and autumn. As it concerns the variability of the MODIS retrievals availability throughout the year, the main driving factor is the clouds since AOD is measured by satellites under clear skies conditions. In the broader Mediterranean basin, the cloud coverage increases from south to north (spatial variability) and from warm to cold seasons (temporal variability). Therefore, both aforementioned reasons (Sun glint, clouds), either solely or combined, can explain the spatial patterns of MODIS-Terra AOD availability displayed in Figures R1-R4 of our previous response. We have modified the relevant part of the manuscript accordingly (Lines: 368-372).

"In brief, although I'm aware the same methodology has been used in several other publications, to my opinion the different data coverage in time and space is a major critical element not properly addressed by the algorithm currently adopted."

See our responses to previous comments.

"[Further comment: good that this unbalance of Figure R1 and R2 somehow 'compensates' in the annual Figure R3 and that this compensation keeps similar for the different years considered (Figure R4). I believe Figure R5 I suggested to produce is indeed interesting and useful. I think the authors should insert it at least in the supplementary material]."

We have added the geographical distributions of the calculated statistics (for both study periods) namely the mean value and the associated standard deviation as well as both threshold levels in the revised supplementary material (Figure S1). A sentence has been added to the manuscript (Lines: 339 - 342) directing the reader to the obtained results.

"Does this mean in METHOD-B the identification of 'pure' DD cases uses the thresholds for Ångström exponent, Fine Fraction, Aerosol Index and Effective radius BUT NO threshold for AOD? The authors should please clarify this point, as the same ambiguity is found in the text added to the revised manuscript (lines 371-383)."

Yes, this is stated clearly in our manuscript (Lines: 399-410). Only the defined criteria for Ångström exponent, fine fraction, effective radius and Aerosol index are used in order to mask out the "pure" dust

AOD retrievals. Then, from the remaining non-dust AOD retrievals the threshold levels (Mean + 2*Std and Mean+4*Std) are calculated for each pixel. Finally, the DD episodes' frequency of occurrence and intensity are calculated based on the defined thresholds.

2. DATASET PROBLEMS

"I understand the authors reasons to keep a different time range for AQUA and TERRA, exploiting the longer TERRA dataset. Still, I think showing the results I asked for (Figures R7 and R8) in the supplementary is important because then (and only then) the authors can substantiate the statement that differences between TERRA and AQUA are likely due to daily cycle effects rather than to the different long-term period addressed."

In the revised supplementary material we have added both figures (Figures S5 and S6).

"A similar text is inserted in the revised manuscript (lines 211-214). Given the 'uncertainties' on the accuracy of these parameters, some 'sensitivity' on the use of the relevant thresholds in the algorithm would have been opportune, as well as on the real need for such a multi-parameter dataset."

In the submitted document (Lines: 366-368) we were stating that the defined thresholds for α, FF, reff and AI have been selected based on raw data availability, literature findings and several sensitivity tests directing the reader to Gkikas et al. (2013). In the revised document, we have added a short description about the obtained results concerning the sensitivity tests (Lines: 391-396).

"This was the information I was asking for in my question. This information needs to be added to the text, together with the explanation on how these 23% and 12.8% percentages are obtained. This information is very interesting, and it is indeed a bit strange no mention to that Gkikas et al. 2016 paper (or manuscript in press) was done in the first version of the manuscript nor in the current revised one (the paper is now published). It should obviously be included in the reference list and mentioned in the introduction to put the present study into a more general prospective."

We have added in the revised manuscript that the algorithm which is used here is a branch of a unified algorithm presented in Gkikas et al. (2016) but we are not discussing other details (available in Gkikas et al. 2016) (See lines 331-335).

"As mentioned in my general comment at the beginning, this manuscript addresses a lot of different aspects of DD events (DD algorithm issues, horizontal scale, vertical scale, comparison to AERONET, comparison to PM10) and seems to lack a main focus. This also translates into a very long and inhomogeneous text that does not help the reading. In my former comment 3.2, I suggested to leave the comparison with PM10 data to a further (interesting) investigation. I'm still convinced of this, and even more now that the revised version further added a 3 pages-section (4.4) on this aspect, with the study of specific 'desert-dust cases'."

We disagree with the Reviewer's proposal for the following reasons. Section 4.4, which has been added in the submitted manuscript following the recommendations made by the Reviewer 3 and the acceptance of the Editor, discusses how the vertical distribution of dust outbreaks affects the level of agreement between columnar (MODIS) and ground (PM) observations. It is apparent that the obtained results (Section 4.4) are directly related to the scientific target of our analysis which is the description of the Mediterranean desert dust outbreaks' vertical structure. In addition, the analysis in Section 4.4 is triggered by the obtained results in Section 4.1.2 where it is investigated the agreement between columnar AODs (MODIS) and ground concentrations (PM) across the Mediterranean. Hence, these two sections are linked together and we believe that should be presented in the manuscript. Moreover, omitting the parts of the analysis related to PM (Sections 4.1.2 and 4.4) and extending those related to the applied methodology would result in an unbalanced manuscript providing little additional information with regards to that of Gkikas et al. (2013).

Some minor comments

"- Some of the information provided in your answers to my minor comments needs to be inserted in the text to improve its clarity and completeness."

We have inserted to the manuscript as many as possible of our responses to the Reviewer's minor comments trying also not to extend further the length of the manuscript (see also our response to the Reviewer's general comment).

"- Declared aim of the study in the abstract is now 'to describe the vertical structure of the intense Mediterranean dust outbreaks' but this is just a part of the work, the horizontal structure over the basin, although less original, is also important within the manuscript and should be also emphasized in the abstract (see also my general comment on the need to better clarify which is the focus of the work and to re-arrange the whole text accordingly)."

The aim of the study, as stated in the first two sentences of the Abstract, is the description of the Mediterranean dust outbreaks and their vertical structure. This already implies that features of their horizontal distribution are examined, as it is immediately understood/realized by the reader when reading about the obtained results of the DD episodes' frequency of occurrence and intensity in the following sentences of the Abstract (lines 32-41). Also note that adding Section 4.4 (specific dust outbreaks) in the revised manuscript resulted in a strengthened part of the analysis related to the vertical extension of dust outbreaks.

"- The authors use the two metrics DD 'episode frequency' and 'intensity' to characterize DD over the Mediterranean. From their description (Section 3), it seems that an 'episode' (per pixel) is a single day of dust, while quite often in the literature the term 'episode' is used for a dust event lasting more than 1 day. This is quite important to clarify, also to allow comparison with other literature data. For example Pey et al., 2013 show a dust frequency (% over annual days) over Southern Europe up to about 37% in Sicily (Italy), and about 30% in Southern Spain and Greece. Note that these percentages in Pey et al. (2013) are obtained ONLY limiting to the cases in which Saharan dust is observed at the ground, thus

should correspond to a subset of the DD statistics from columnar measurements as in the present work. Here Gkikas et al get maximum frequencies for 'strong' events of about 10 episodes/year (i.e. less than 3% over annual days) close to the African coasts. How do the authors reconcile these numbers? Could they please comment on that? Note that this aspect is somehow connected to the definition of the 'AOD thresholds', from which all the subsequent analysis and definition of 'strong' and 'extreme' events derives."

We agree with the Reviewer that in literature the term "episodes" usually refers to unusually high concentrations recorded persistently for consecutive days but this does not mean that it cannot be used also for single days, as defined in the present and our previous papers. Yet, and based on this definition, the duration of DD episodes is easily understood and quantified, as done in Gkikas et al. (2013). We have added a brief clarification in Lines: 95-96. As it concerns the second part of the Reviewer's comment, we would like to mention that it is normal to obtain different numbers (frequencies) because of the different approaches taken in different studies, as stated by the Reviewer himself/herself. In Pey et al. (2013), dust occurrences are identified when dust is recorded at stations regardless its concentration while in our analysis we are identifying intense dust episodes when AOD levels are higher/equal than mean plus two times standard deviation. Hence, these differences are attributed to the different "thresholds" definition. We have added the relevant information in the revised document (Lines: 685-691).

"- Figure 1: The authors can keep it if adding a second panel showing the equivalent scheme for the METHOD-B, this making the figure different from the one in their former publication."

We have decided to remove Figure 1 from the revised manuscript following the initial suggestion made by the Editor.

"- Figures 9 and 10 are not very much readable and should be improved."

We have increased furthermore the size of Figures 9 and 10.

"- I did not have the opportunity to fully read the Gkikas et al (2016) paper on Atmospheric Environment, but why are the Figures 2 and 3 in this manuscript different from Figures 4 and 5 in that paper (for those plots referring to the same DD analysis on the same MODIS Terra dataset)? It would also be important to comment on these differences, given that this manuscript follows that other one from the authors."

The analysis in Gkikas et al. (2016) covers the period Mar. 2000 – Feb. 2007 while in the present paper the study period extends from March 2000 to February 2013. Moreover, in Gkikas et al. (2016) the satellite algorithm operates with non-weighted QA retrievals in contrast to the present analysis (QA weighted). For the aforementioned reasons, the obtained results from both studies are not identical. Note that the DD episodes presented in Gkikas et al. (2016) are obtained with the same previous version of algorithm utilized in Gkikas et al. (2013). Therefore, in this study we discuss the differences between the present analysis (improved algorithm) and Gkikas et al. (2013, previous algorithm).

"- I noticed the following reference is reported twice in the reference list, with two different years of publication:

- Gkikas, A., Houssos, E. E., Lolis, C. J., Bartzokas, A., Mihalopoulos, N. and Hatzianastassiou, N.: Atmospheric circulation evolution related to desert-dust episodes over the Mediterranean. Q.J.R. Meteorol. Soc., 141: 1634–1645. doi: 10.1002/qj.2466, 2015.

- Gkikas, A., Houssos, E. E., Lolis, C. J., Bartzokas, A., Mihalopoulos, N. and Hatzianastassiou, N.: Atmospheric circulation evolution related to desert-dust episodes over the Mediterranean. Q.J.R. Meteorol. Soc., doi: 10.1002/qj.2466, 2014."

We would like to thank the Reviewer for noticing our mistake. In the revised manuscript we have kept the first reference which is the correct one.

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° x 1° 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

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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*=0.505-0.750) 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 32º N and 38º 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 km^{-1} 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 (~ 0.004 km^{-1} 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).

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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; Solmon et al., 2008). Moreover, it has been shown that the consideration of their radiative impacts in numerical simulations can improve the forecasting accuracy of weather models (Pérez et al., 2006). Dust particles also affect air quality in urban areas (Basart et al., 2012) causing adverse health effects (Díaz et al., 2012; Karanasiou et al., 2012; Pérez García-Pando et al., 2014). All these consequences of dust aerosol are relevant and maximize under maximum dust loads, namely dust episodes, highlighting thus the significance of analyzing the spatial and temporal characteristics of such events. To this aim, many studies have been carried out using either surface (e.g. Cachorro et al., 2006) or satellite (e.g. Moulin et al., 1998) observations, as well as modelling techniques (e.g. Heinold et al., 2007) focusing on the broader Mediterranean area. These studies have been done either for specific cases (e.g. Kubilay et al., 2003; Balis et al., 2006) or for extended periods at specific locations (e.g. Meloni et al., 2007; Toledano et al., 2007a; Gobbi et al., 2013; Mona et al., 2014). Recently, Gkikas et al. (2013) developed an objective and dynamic algorithm relying on satellite retrievals, which enabled an overall view of dust episodes over the entire Mediterranean and the characterization of their regime (i.e., frequency of occurrence, intensity and duration).

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

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

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

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

Surface-based lidar measurements like those used in the aforementioned studies provide useful information about the geometrical and optical properties of dust layers, but they are representative only for specific locations. Yet, a more complete knowledge about the vertical structure of dust outbreaks is necessary in order to adequately understand and determine their possible effects. The limitation imposed by the use of surface-based lidar observations can be overcome by utilizing accurate satellite retrievals, as a complementary tool, which provide extended spatial coverage. Since 2006, vertical resolved observations of aerosols and clouds from space were made possible thanks to the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) lidar flying onboard the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite (Winker et al., 2009). Based on CALIOP observations, Liu et al. (2008) analyzed the global vertical distribution of aerosols for one year, while other studies focused on the vertical structure of dust outflows towards the Atlantic Ocean (e.g. Ben-Ami et al., 2009; Adams et al., 2012; Tsamalis et al., 2013) and the Pacific Ocean (e.g. Eguchi et al., 2009; Hara et al., 2009). On the contrary, over the broader Mediterranean area, only a small number of studies has been made aiming at describing the vertical distribution of dust aerosols (Amiridis et al., 2013) or specifying the vertical structure of dust events (Amiridis et al., 2009). Nevertheless, they only dealt with a single dust event (18-23 May 2008, Amiridis et al., 2009) and thus cannot satisfy the need to know the general vertical structure of Mediterranean dust episodes.

The main target of the present study is to describe the Mediterranean desert dust outbreaks' vertical structure. For this purpose, satellite retrievals derived by the MODIS-Terra/Aqua, ~~Earth Probe~~EP-TOMS, OMI-Aura and CALIOP-CALIPSO databases (Section 2) are used in a synergistic way. The dust outbreaks are identified with an objective and dynamic algorithm, which uses appropriate aerosol optical properties representative of suspended particles' load, size and nature (Section 3). First, ~~T~~the outputs of the ~~default version of the~~ satellite algorithm are compared versus surface measurements provided by AERONET or *PM_{10}* stations, located within the study region (Section 4.~~2~~1). Additionally, useful information about various optical and physical properties under intense dust episodes conditions is also derived from the aforementioned analysis. Then, ~~Based on its outputs,~~ the primary characteristics of the intense Mediterranean desert dust (*DD*) episodes, namely their frequency and intensity, are described in Section 4.~~1~~2. Just in order to assess the consistency of the algorithm' concept, an alternative methodology for the determination of *DD* episodes is also applied and the obtained results are inter-compared with the basic methodology. ~~The outputs of the default version of the satellite algorithm are compared versus surface measurements provided by AERONET or PM_{10} stations, located within the study region (Section 4.2). Additionally, useful information about various optical and physical properties under intense dust episodes conditions is also derived from the aforementioned analysis.~~ For the identified *DD* episodes, collocated CALIOP-CALIPSO vertical feature mask and total backscatter coefficient at 532 nm retrievals are used in order to describe the annual and seasonal variability of dust outbreaks' vertical extension over the Mediterranean (Section 4.3). Moreover, in Section 4.4, a thorough analysis of ~~few~~ specific Mediterranean *DD* episodes is made, in order to examine how the vertical distribution of desert dust outbreaks can affect the agreement between MODIS *AOD* and *PM_{10}* data. Finally, the summary and conclusions are drawn in Section 5.

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2. Satellite and surface-based data

The different types of satellite retrievals that have been used as inputs to the objective and dynamic satellite algorithm are described below, namely the MODIS (Section 2.1.1), EP-TOMS and OMI-Aura

(Section 2.1.2) databases. Also, CALIOP-CALIPSO vertically resolved satellite data, coincident with the identified desert dust outbreaks by the satellite algorithm, are described in Section 2.1.3. Finally, surface-based sun-photometric AERONET retrievals and PM_{10} concentrations, both used for the comparison against the satellite algorithm's outputs, are described in Sections 2.2.1 and 2.2.2, respectively.

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2.1 Satellite data

2.1.1 MODIS

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

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The following daily MODIS-Terra and MODIS-Aqua Collection 051 (C051) level 3 satellite data (MOD08_D3 and MYD08_D3 files) provided at 1° × 1° latitude-longitude spatial resolution are used: (i) AOD_{550nm}, (ii) Ångström exponent over land ($\alpha_{470-660nm}$), (iii) Ångström exponent over ocean ($\alpha_{550-865nm}$), (iv) fine-mode fraction (*FF*) of *AOD* over land and ocean and (v) Effective radius over ocean (r_{eff}). It must be mentioned that the size parameters (*α*, *FF*) over land are less reliable compared to the corresponding ones over sea, since they are highly sensitive to spectral dependent factors such as errors in the surface model or sensor calibration changes. Over sea, the accuracy of size parameters is strongly dependent on wind conditions.

Similar data have been used by Gkikas et al. (2013), however, in the present study we have improved data quality by using the quality assurance-weighted (QA) level 3 data (http://modis-atmos.gsfc.nasa.gov/_docs/QA_Plan_2007_04_12.pdf) derived from the level 2 retrievals (10 km x 10 km spatial resolution). Each level 2 retrieval, is flagged with a bit value (from 0 to 3) corresponding to confidence levels (No confidence: 0, Marginal: 1, Good: 2 and Very Good: 3). Based on this, the level 3 QA-weighted spatial means are obtained by the corresponding level 2 retrievals considering as weight their confidence level (bit value). In addition, the day cloud fraction as well as the number of level 2 counts, which are both relevant to the performance of the satellite algorithm, are also used in this study. The time series of daily MODIS aerosol data cover the 13-yr period March 2000-February 2013 (Terra) and the 10-yr period January 2003-December 2012 (Aqua).

2.1.2 EP/TOMS and OMI-Aura

The selected retrievals from MODIS provide information about particles' load (*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-

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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).

2.1.3 CALIOP-CALIPSO

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

In the present analysis, we use the Version 3 (3.01 and 3.02) of the Level 2 Vertical Feature Mask (*VFM*) and Aerosol Profile Products (*APro*) files, available from June 2006 to February 2013, both derived from the NASA's Earth Observing System Data and Information System (http://reverb.echo.nasa.gov/). The aerosol profile products are generated at a uniform horizontal resolution of 5 km (http://www-calipso.larc.nasa.gov/products/CALIPSO_DPC_Rev3x6.pdf), while the vertical resolution varies from 60 to 180 m depending on the altitude range and the parameter. The scientific data sets which have been analyzed are the following: (i) aerosol subtype, (ii) *CAD* score and (iii) Total Backscatter Coefficient at 532 nm (β_{532nm}), reported at several tropospheric and stratospheric levels above mean sea level (Hunt et al., 2009).

2.2 Surface-based data

2.2.1 AERONET

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

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2.2.2 PM₁₀

Daily total and dust surface *PM₁₀* concentrations, over the period 2001-2011 from 22 regional background and suburban background sites were used in this study. The monitoring sites are distributed as follows: 10 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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3. Identification of desert dust episodes

~~Following the methodology proposed by Gkikas et al. (2013), desert dust (*DD*) episodes are identified based on an objective and dynamic algorithm (see Figure 2 in Gkikas et al., 2013) which consists a branch of a unified algorithm able to identify not only *DD* episodes, but also four other types of aerosol episodes, namely biomass-urban (*BU*), dust/sea-salt (*DSS*), mixed (*MX*) and undetermined (*UN*). The unified algorithm has been applied by Gkikas et al. (2016) in order to characterize aerosol episodes in the greater Mediterranean Sea area, over the period Mar. 2000 – Feb. 2007.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.~~

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~~The present ("dust") algorithm operatesThe 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).~~ Following the methodology proposed by Gkikas et al. (2013), desert dust (*DD*) episodes are identified based on an objective and dynamic algorithm which consists a branch of a unified algorithm (Gkikas et al., 2016) able to identify and characterize not only *DD* episodes, but also four other types of aerosol episodes, namely biomass-urban (*BU*), dust/sea-salt (*DSS*), mixed (*MX*) and undetermined (*UN*). The algorithm (see Figure 2 in Gkikas et al., 2013) 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*. The geographical distributions of the computed statistics (*Mean* and *Std*) as well as the corresponding spatial patterns of both threshold levels are displayed in Figures S1-a (MODIS-Terra, Mar. 2000 – Feb. 2013) and S1-b (MODIS-Aqua, 2003 – 2012) in the supplementary material. At the next step, the algorithm analyzes the daily AOD_{550nm} timeseries and classifies an episode as a strong one when *AOD* is between the two defined thresholds ($Mean+2*Std \leq AOD_{550nm} < Mean+4*Std$) and as an extreme one when *AOD* is higher/equal than *Mean+4*Std* ~~(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 *AODs*, also possibly relevant to radiative and health effects, are masked out from the dataset. In order to investigate the possible impact of this, "unbiased" mean, standard deviation and thresholds of *AOD* are also computed based on another methodology and the results are discussed comparatively to those of the primary methodology in a separate paragraph. Moreover, it must be mentioned that the satellite algorithm identifies only intense desert dust episodes since their *AOD* must be higher than *Mean+2*Std* which is considered as a high threshold level.

It should be noted that the representativeness of the calculated mean levels is possibly affected by the availability of the *AOD* retrievals and particularly by the way these data are distributed both at temporal and spatial ~~different temporal~~ scales. Thus, a possible underrepresentation of winter *AOD* data in the long-term dataset, which is often the case in satellite retrievals of *AOD*, may result in a smaller mean *AOD* than what would be in case of complete and balanced seasonal availability. ~~It should be noted that t~~Moreover, the spatiotemporal availability of *AOD* is determined by the different satellite retrieval algorithm assumptions depending on the underlying surface type (land or sea) and clouds (i.e. satellite retrievals are possible only under clear skies conditions). In order to investigate the possible effect of temporal availability of daily *AOD* data~~is,~~ ~~To this aim~~, we have calculated the percentage

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availability of *AOD* retrievals on a monthly, seasonal and year by year basis, over the period 2000-2013 (results not shown here). Seasonal differences of *AOD* availability are mainly encountered in the northernmost parts of the study region, attributed to the enhanced cloud coverage, with lower values (20 to 40 %) from December to February against 50-85% for the rest of the year. Differences of *AOD* availability ~~B~~are also found between land and sea surfaces ~~are also found differences of *AOD* availability~~ which are more pronounced in winter and summer and less remarkable during the transition seasons. More specifically, across the Mediterranean Sea, in winter, the availability percentages range from 70 to 90 % while in summer the corresponding values are decreased, due to Sun glint, down to 60 % and 80 %, respectively. Over land, for both seasons, the spatial patterns of *AOD* availability are reversed. ~~This is attributed to the enhanced cloud coverage prohibiting the satellite observations.~~ In order to investigate furthermore how the spatiotemporal *AOD* variability and unbalanced seasonal distribution of MODIS *AOD* data can affect the calculated mean *AOD* levels (calculated by daily retrievals) we have repeated the calculations by utilizing monthly retrievals (calculated by the daily ones) thus removing the unequal seasonal contribution to the long-term mean AOD values. According to our results, only small differences are found, generally hardly exceeding 0.1~~-0.2~~ ~~0.2~~ in absolute and 5% in relative percentage terms, with the mean *AODs* over land ~~are~~being higher by up 10 % when th~~ese~~ey are computed from daily than monthly data, while the opposite is found over sea. This finding reveals ~~thus~~ that the unequal temporal distribution of *AOD* retrievals does not have critical impact on the computed mean *AODs* and the resulting algorithm outputs presented in this study. ~~The loss of satellite *AOD* retrievals in winter can affect the statistics and computed long-term *AOD* mean values and associated thresholds. According to performed computations, such losses of up to 70% daily values in winter months result in overestimated *AOD* thresholds by up to 4% and 3% for strong and extreme DD episodes. This is small overestimation does not affect essentially the outputs of the present algorithm, namely the computed frequency and intensity of DD episodes. The 4% and 3% are the maximum possible uncertainties of the algorithm which are encountered in the northernmost parts of the study region (e.g. Balkans), whereas in the rest of the area corresponding uncertainties are even much smaller. Nevertheless, this does not essentially affect the algorithm outputs since these regions, being far away from the dust sources, are not so frequently affected by dust outbreaks, especially given the significant wet removal of aerosols during this most rainy season of the year. On a year by year basis, the differences of the *AOD* data's availability are almost negligible.~~

In a further step of the methodology, the strong and extreme *DD* episodes are identified separately over land and sea surfaces of the study region. This is achieved through the usage of specific aerosol

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optical properties, namely the Ångström exponent, effective radius, fine fraction and aerosol index, which provide information about particles' size and nature (black box, Figure 1). For each optical property, appropriate upper or lower thresholds have been set up (green boxes, Figure 1) which must be valid concurrently in order to certify the presence of dust particles in the atmosphere. Note that there are not any unanimously defined acknowledged thresholds in literature. Therefore, tThese cut-off levels have been selected here according to the literature findings, availability of raw data and several own sensitivity tests (more details are provided in Gkikas et al., 2013) which have been applied individually to the MODIS size parameters (i.e., *α*, *FF* and r_{eff}). Such analysis is essential when multi-parameter datasets are utilized and their variations can possibly modify the satellite algorithm's outputs. To this aim, we have applied the satellite algorithm alteringmodifying by 0.1 the *α*, *FF* and r_{eff} values forwithin the ranges 0.6 – 0.8, 0.1 – 0.4 and 0.4 – 0.8, respectively. Our results indicate that the geographical patterns remain similar and the total number of *DD* episodes is only slightly modified (less than 4 %) for the *α* and r_{eff} retrievals, whereas it changes more for the FF retrievals (by up to 25% over sea for strong episodes). On the contrary, the highest sensitivity is found for the *FF* retrievals over sea where *DD* episodes, particularly the strong ones, are decreased abruptly (> 25 %) even for a small decrease (0.1) of the *FF* cut-off level. –Here, Tthe validity of these thresholds is further evaluated against AERONET measurements and the corresponding results are discussed in Section 4.21.1.4.

In order to address the issue of possible overestimation of the defined threshold levels, particularly in the most dust affected areas as it has been mentioned above, we have also applied the satellite algorithm using an alternative methodology (METHOD-B) in which dust-affected grid cells were excluded. In this case, from the raw *AOD* retrievals we have masked out the "pure" desert dust grid cells, which were identified based on the concurrent fulfillment of the defined criteria for dust occurrence in the algorithm (for Ångström exponent, fine fraction, aerosol index and effective radius, green boxes of Figure 1). Then, from the remaining data (non-dust *AOD* retrievals), the mean, the associated standard deviation as well as the defined thresholds of *AOD* are computed for the whole study period, for each pixel, as also done in the primary methodology. Finally, also similarly to the way done in the primary methodology, the *DD* episodes were classified into strong and extreme ones. The obtained results, i.e. frequency of occurrence and intensity of *DD* episodes, based on the primary methodology and METHOD-B are discussed in Section 4.2.The frequency of occurrence and intensity of *DD* episodes determined with METHOD-B are provided in the supplementary material (Figures S12 and S23) while their differences with regards to the primary methodology are discussed in Section 4.12.

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

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4. Results

Before dealing with the horizontal patterns (sub-section 4.2) and the vertical structure of dust outbreaks (sub-sections 4.3 and 4.4), it is very important to ~~describe their horizontal patterns (sub-section 4.1) and also to~~ compare the algorithm's outputs against quality AERONET and PM_{10} observations (sub-section 4.~~2~~1) in order to ensure an accurate three-dimensional view of the intense Mediterranean *DD* episodes. It must be clarified, that the comparison of the satellite algorithm's outputs versus AERONET/PM_{10} is made only for its default version and not for the METHOD-B, since between the two methodologies are not found remarkable differences, as it will be presented in Section 4.~~1~~2. ~~Accordingly~~For the same reason, the synergistic implementation of the 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.

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4.~~2~~1 Comparison of the satellite algorithm's outputs against AERONET and PM_{10} measurements

The ability of the satellite algorithm to identify satisfactorily *DD* episodes, is tested against ground measurements from 109 AERONET (Fig. ~~4~~1, orange squares) and 22 PM_{10} (Fig. ~~4~~1, green triangles)

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stations located in the broader Mediterranean area. This is an extended and thorough comparison which exceeds largely a similar one done for the outputs of the previous version of satellite algorithm (2000-2007, Gkikas et al., 2013), but only relying on 9 AERONET stations and using *AOD* and volume size distribution data. Here, the comparison is repeated for the improved algorithm, being extended over a longer time period, for a much larger number of AERONET stations, and an analysis of more optical properties, namely the Ångström exponent, effective radius, single scattering albedo and asymmetry parameter is made. The comparison is performed for both study periods and satellite platforms (Mar. 2000 – Feb. 2013 for Terra and 2003–2012 – 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 41, 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 41) the intense *DD* episodes have been identified from ground (AERONET) and the corresponding results are compared with the satellite algorithm outputs (Section 4.21.1.4). Finally, the performance of the algorithm is also tested against surface PM_{10} measurements from 22 stations (Section 4.21.2).

4.21.1 AERONET

4.12.1.1 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. The selected dataset corresponds to 1.06 % of the overall (strong and extreme) *DD* episodes (32635) which have been identified during the study period. It should be noted that AERONET

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AOD_{550nm} values have been calculated from available AERONET AOD_{870nm} and Ångström exponent data ($\alpha_{440\text{-}870nm}$) 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~~2. Two similar scatterplots with matched MODIS-AERONET data pairs are given. The first one (Fig. ~~5~~2 i-a) is resolved by the number of level 2 (*L2*) measurements of 10 km x 10 km spatial resolution from which the compared 1^{o} x 1^{o} level 3 (*L3*) *AODs* in the figure are derived. The second scatterplot (Fig. ~~5~~2 i-b) is resolved by the spatial standard deviation inside the 1^{o} x 1^{o} geographical cell (level 3 *AODs*). Both scatterplots address the issue of level 3 *AOD* sub-grid spatial variability, which is essential when attempting comparisons against local surface-based *AOD* data like the AERONET.

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

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

~~In addition~~Finally, the spectral variation of the AERONET *AODs* at 7 wavelengths, from 340 to 1020 nm, in climatological and dust episodes conditions has been investigated (results given in Figure ~~S3~~S2, supplementary material). The *AOD* boxplots produced for all the available daily AERONET measurements (orange) and for the corresponding retrievals during strong (cyan), extreme (red) and all *DD* (green) episodes identified by the satellite algorithm show that the spectral variation of aerosol optical depth decreases in cases of dust episodes, with respect to the "climatological" conditions. This is mainly attributed to the further increasing *AOD* levels at wavelengths longer than 500 nm (by about 6 times) than in (or near) the visible.

4.~~2~~1.1.2 Aerosol volume size distribution

In Figure ~~6~~3, are presented the mean aerosol volume size distributions (*AVSDs*) calculated from all available AERONET data (orange curve) as well as under strong (cyan curve), extreme (red curve) and all (green curve) *DD* episodes conditions. The results are given for Mar. 2000 – Feb. 2013 using MODIS-Terra (346 intense *DD* episodes) retrievals as inputs to the satellite algorithm. In the climatological curve, two modes are distinct centered at 0.15 μm for the fine mode and 2.24 μm for the coarse mode. There is an about equal contribution of both modes, indicating the coexistence of fine (e.g. urban aerosols) and coarse (e.g. dust aerosols) particles over the broader Mediterranean area. This result is in agreement with previous studies for the Mediterranean (e.g. Fotiadi et al., 2006; Mallet et al., 2013). However, under dust episodes conditions, although the *AVSD* still has two modes, there is a dramatic increase of the coarse mode, which strongly dominates. More specifically, the peak of the coarse mode (radius between 1.7 and 2.24 μm) is increased by factors of about 10, 15 and 11 for the strong, extreme and all *DD* episodes. The differences between the strong and extreme *AVSDs* are statistically significant (confidence level at 95 %) for almost all size bins (18 out of 22) except bin 1 (0.050 μm), 2 (0.065 μm), 6 (0.194 μm) and 7 (0.255 μm). Moreover, it should be noted that the increment factors are slightly decreased when the algorithm operates only with *AODs* associated with cloud fractions less than 0.8 which is reasonable since possible "overestimated" retrievals are masked

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out from the analysis. Similar modifications in the shape of *AVSD* during dust outbreaks have been pointed out by several studies in the past, either for the Mediterranean region (e.g. Kubilay et al., 2003; Lyamani et al., 2005; Córdoba-Jabonero et al., 2011) or for other dust affected areas of the planet (e.g. Alam et al., 2014; Cao et al., 2014).

4.~~2~~1.1.3 Size optical properties, single scattering albedo and asymmetry parameter

The accuracy of the *DD* episodes identification method was further assessed by also using other AERONET aerosol optical properties than *AOD*, namely the Ångström exponent (α) and the effective radius (r_{eff}), able to provide information about particles' size. For both aerosol optical properties, the boxplots for all the available AERONET retrievals as well as for the corresponding data during strong, extreme and all *DD* episodes, have been produced and depicted in Figure ~~S4~~ S36 (supplementary material).

Based on our results, the appropriateness of the applied methodology is confirmed by the drastic reduction of α and increase of r_{eff} values when dust outbreaks occur. When all available AERONET 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 (Figure S3-i). On the contrary, during intense dust episodes the majority of the corresponding values for all and strong *DD* episodes are lower than 0.54 while for the extreme ones are lower than 0.36. Such low Ångström exponent values, attributed to transported mineral particles from the northern African deserts (Pace et al., 2006), have been reported also in previous studies (e.g. Tafuro et al. 2006; Basart et al., 2009). The existence of coarse aerosols is also confirmed by the increase of r_{eff} values under intense *DD* conditions compared to the climatological levels (Figure S3-ii). 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 S~~5~~74-i) and g_{aer} (Figure S~~5~~74-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~~09,

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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~~1.1.4 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~~1, during the periods for which ground retrievals are available (Table 1). The selection of the AERONET stations was based on: (i) data availability (see last column of Table 1), (ii) their location (i.e. near to the Northern African and Middle East deserts) and (iii) the inclusion of sites where the aerosols' regime is complex (e.g. El Arenosillo, FORTH Crete). The intense *DD* episodes were identified following the methodology described in section 3, but using only *AOD* at 870 nm, $\alpha_{440\text{-}870nm}$ (lower/equal than 0.7) and r_{eff} (higher than 0.6) as criteria, based upon their availability from AERONET. Subsequently, the algorithm was also operated again using satellite (MODIS-Terra, OMI-Aura, EP-TOMS) input data for the days with available retrievals in each of the 7 AERONET stations.

In Figure ~~7~~4, we present the overall scatterplots between satellite and ground *AODs* when intense *DD* episodes have been identified based on the ground (left column) and the satellite (right column) algorithm. Colors in Figs. ~~7~~4 i-a, ~~7~~4 ii-a, ~~7~~4 iii-a represent the associated MODIS-Terra Ångström exponent, effective radius and day cloud fraction~~ (*CFD*)~~ retrievals, respectively. In Figs. ~~7~~4 i-b and ~~7~~4 ii-b colors represent the AERONET Ångström exponent and effective radius, respectively, while in Figure ~~7~~4 iii-b represent the day cloud fraction observations derived by MODIS-Terra. Through this approach it is feasible to assess furthermore the performance of the satellite algorithm, specify its drawbacks and check the validity of the defined thresholds (green boxes in Figure ~~1~~2 in Gkikas et al., (2013)).

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It is apparent that the agreement between MODIS-Terra and AERONET *AODs* is better when *DD* episodes are identified from the ground, as shown by the increased correlation coefficients (from 0.521 to 0.704), increased slopes (from 0.6 to 0.9-1.0) and decreased biases (from 0.16 to -0.03). In particular, when *DD* episodes are identified from space, the MODIS-Terra *AOD* retrievals are overestimated (bias=0.163) with regards to AERONET, particularly at low *AOD* values (< 0.5). In both algorithms, the highest overestimations are associated with cloud fractions higher than 0.7 due to the possible contamination of the satellite *AODs* by clouds (Figure ~~7~~ 4 iii-a, iii-b). Given that *DD* episodes' identification based on AERONET retrievals is more efficient, we have used these results in order to check the validity of the defined thresholds for *α*, *AI*, *FF* and r_{eff} ~~(green boxes in Figure 1)~~ used in the satellite algorithm. For each aerosol optical property, it has been calculated the percentage of intense *DD* episodes for which the corresponding satellite observations are below or above the defined thresholds, depending on the parameter. The results given in Table 2 are satisfactory, since the percentages range from 87 to 99%, and confirm the validity of the defined thresholds.

The scatterplots in Figs. ~~7~~4 i-b and ii-b also reveal some weaknesses of the satellite-based algorithm. More specifically, it is found that for few *DD* episodes identified by the satellite algorithm the corresponding AERONET Ångström exponent and effective radius values are higher than 1 and smaller than 0.4, respectively. These values indicate a predominance of fine particles instead of coarse ones as it would be expected 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 also attributed to the lower accuracy of MODIS aerosol size retrievals over land (Section 2.1.1).

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4.~~2.2 1~~1.2 PM_{10} and dust contribution

The satellite algorithm's outputs, apart from AERONET retrievals, have been also compared against ground PM_{10} concentrations (μg m^{-3}) measured in 22 Mediterranean stations (green triangles in Figure ~~4~~1).

First, for each station, the number of intense *DD* episodes was calculated, for which coincident satellite and ground measurements (total PM_{10}) are available (Figure ~~8~~5-i). The number of concurrent *DD* episodes varies from 3 to 53, being in general decreasing from southern to northern stations. For 14 out of 22 stations, where at least 10 intense *DD* episodes were identified by the satellite-based algorithm, we have computed the correlation coefficients between satellite *AODs* and surface total PM_{10} concentrations (Fig. ~~8~~5-ii). The highest *R* values (up to 0.8) are recorded in the central and eastern parts of the Mediterranean while the lowest ones are found in the western stations. It must be noted that the correlation coefficients are affected by outliers, because of the limited number of *DD* episodes in each station, highlighting the sensitiveness of the intercomparison. Such outliers can be expected when satellite-based columnar *AODs* and surface-based PM_{10} data are compared, since satellite *AODs* are representative for the whole atmospheric column in contrast to in-situ *PM* measurements which are more representative for the lowest part of the planetary boundary layer affected also by local factors. Therefore, the vertical distribution of desert dust load, as it will be presented in the next sections, can determine the level of agreement between satellite *AODs* and surface *PM* concentrations. ~~Another influencing factor can be cloud contamination of MODIS *AOD*.~~

The identification method by the satellite algorithm can be considered as correct when dust PM_{10} concentrations are higher than zero (i.e. dust has been recorded at the station). According to this, the ratio between the number of non-zero dust *PM* observations and the number of *DD* episodes (coincident satellite-derived *DD* episodes and total PM_{10} measurements) for each station is defined as success score. The calculated success scores (Figure ~~8~~5-iii) vary from 68% (Monagrega, northeastern Spain, 28 episodes) to 97% (Boccadifalco, Sicily, 33 episodes) confirming the appropriateness of the *DD* episodes' identification. In the majority of stations, the contribution of dust particles to the total burden (Figure ~~8~~5-iv) is above 50%, ranging from 44% (Zarra, Spain) to 86.8% (~~Ayia~~ Agia Marina, Cyprus). In order to complete our analysis we have also calculated the mean (Figure ~~8~~5-v) and the median (Figure ~~8~~5-vi) dust PM_{10} concentrations for the identified intense *DD* episodes in each station. The mean PM_{10} concentrations mainly vary between 20 and 50 μg m^{-3}, being higher in the southern stations, as expected. The minimum mean value (17 μg m^{-3}) was recorded in Censt (Sardinia) and the maximum one (223 μg m^{-3}) in A~~y~~gia Marina (Cyprus). Our values are much higher than the corresponding ones in Querol et al. (2009b), who obtained that the mean levels of mineral matter in PM_{10} during dusty days range from 8 to 23 μg m^{-3} based on ground concentrations derived by 21 Mediterranean stations. These differences are reasonable since here only intense desert dust outbreaks associated with high aerosol optical depths are considered. Finally, the median PM_{10} concentrations are lower compared to the average ones, indicating that outliers (cases with extremely high *AOD* or PM_{10}) can alter the results, attributed to the fact that both parameters' (*AOD* and PM_{10}) distributions are not Gaussians. For this reason the highest differences are found in Finokalia (Crete) and Agia Marina (Cyprus), where the maximum daily PM_{10} concentrations, equal to 690 and 1291 μgm^{-3}, respectively, were recorded during an intense dust outbreak affected the eastern Mediterranean on 24 and 25 February 2006.

4.~~1~~2 2D geographical distributions of desert dust episodes' frequency and intensity

The mean geographical distributions of strong and extreme *DD* episodes' frequency of occurrence (episodes yr^{-1}) are presented in Figure ~~2~~6. Results are given separately as obtained from MODIS-Terra and Aqua for the periods Mar. 2000 – Feb. 2013 and 2003 – 2012, corresponding to local late morning-to-noon (Terra) and afternoon (Aqua) conditions, respectively. It is evident a gradual reduction of frequencies from south to north, while for the strong *DD* episodes also appears a west to east decreasing gradient. The decreasing south-to-north gradient of intense *DD* episodes' frequency, which is also in agreement with previous studies based on ground *PM* measurements (Querol et al., 2009b; Pey et al., 2013), model simulations (Papayannis et al., 2008; 2014) and AERONET *AOD* retrievals (Basart et al., 2009), can be attributed to the increasing distance from the major dust sources and to the higher precipitation amounts at the northern parts of the basin (e.g. Marrioti et al., 2002; Mehta and Yang, 2008).

The maximum frequencies (9.9 episodes 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. Our calculated frequencies are significantly lower than the corresponding ones obtained in Pey et al. (2013), who studied the African dust intrusions towards the Mediterranean basin, based on ground *PM* concentrations, over the period 2001 – 2011. The observed de~~clinations~~viations between the two studies are mainly attributed to the different thresholds definition and hence nature of dust episodes. Here, focus is given on the intense dust outbreaks (intensity equal/higher than ~~*AOD*~~*Mean* + *2*Std*) while in Pey et al. (2013) the dust occurrences were identified even at very low concentrations (> 1 μg m^{-3}). ~~Apart from the threshold definitions, other reasons which can contribute to these aforementioned discrepancies are that the satellite retrievals are not continuous, in contrast to *PM* measurements, as well as that from columnar retrievals (e.g. MODIS) the identification of "pure" dust conditions, particularly in areas like the Mediterranean where different aerosol types coexist, in many cases is not feasible.~~

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~~ – 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~~Figures S~~34~~5 and S~~45~~6 in the supplementary material), 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~~ – 2012, revealing a constant dust episodes' regime. Therefore, the discrepancy appeared between MODIS-Terra and MODIS-Aqua spatial distributions, is attributed to the diurnal variation of factors regulating the emission and transport of dust particles from the sources areas. Schepanski et al. (2009), analyzed the variation of the Saharan dust source activation throughout the day, based on MSG-SEVIRI satellite retrievals, reporting that dust mobilization is more intense in the local early morning hours after sunrise. Note, that desert dust episodes over the period Mar. 2000 – Feb. 2013 have been identified based on observations retrieved by the Terra satellite, which flies over the study region around noon in contrast to Aqua which provides aerosol measurements at early afternoon hours.

The analysis has been also repeated (results not shown here) considering as inputs to the satellite algorithm only *AODs* associated with cloud fractions lower/equal than 0.8, in order to investigate possible modifications to our results ~~(Figs 2 and 3)~~ due to the cloud contamination effect. As it concerns the strong *DD* episodes, the geographical distributions are similar with those of Fig. ~~2~~6, but

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

The maps of intensities (in terms of AOD_{550nm}) of *DD* episodes (Figure ~~3~~7), show that for both study periods and satellite platforms, the maximum intensities are over the Gulf of Sidra and the Libyan Sea, along the northern African coasts. These intensities reach *AODs* up to about 1.5 for strong and 4.1 for extreme episodes, while the minimum ones (values down to 0.25-0.46) are recorded in the northern and western Mediterranean parts. Note that dissimilar spatial patterns appear between the geographical distributions of *DD* episodes' frequency and intensity, indicating that these two features are determined by different factors (e.g. tracks or strength of depressions). Finally, when the cloud contamination is minimized using only *AODs* associated with *CF* lower than 0.8, then the maximum intensities are shifted southwards, across the northern Africa and eastern coasts of the Mediterranean, being lower than 1 and 2 for strong and extreme *DD* episodes, respectively. Through the rejection of possibly overestimated *AODs* from the dataset, it is found that the threshold levels are decreased (mainly over the most frequently dust affected areas) since both mean and standard deviation values are lower (results not shown here). Nevertheless, even though these *AODs* can be overestimated, in the majority of the cases the collocated AERONET *AODs* are high (but lower than the satellite observations) indicating the occurrence of desert dust outbreaks as it ~~will~~ has be shown in Section 4.~~2~~1.1.4.

The analysis has been also repeated applying the alternative METHOD-B described in Section 3. Just to ensure a longer temporal coverage, this analysis was done for the period Mar. 2000~~-~~ - Feb. 2013~~Feb. 2013~~ using MODIS-Terra data. The obtained results for the frequency of occurrence as well as for the intensity of *DD* episodes are depicted in Figures ~~S1~~ S7 and ~~S2~~S8, respectively, in the supplementary material. The geographical patterns for the frequency of occurrence between the two methodologies are similar; however, the maximum values for the strong and extreme *DD* episodes can reach up to 13.3 episodes $year^{-1}$ (Fig. ~~S1~~S7-i) and 8.1 episodes $year^{-1}$ (Fig. ~~S1~~S7-ii), respectively. As it concerns the intensity, the geographical patterns, particularly for the strong *DD* episodes, are dissimilar and less distinct compared to the corresponding ones obtained with the primary methodology. This difference is attributed to the inclusion of more dust episodes with variable intensity, which leads to a

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not so clear "signal" when all these episodes are averaged. Based on METHOD-B, the maximum intensities (in terms of AOD_{550nm}) of strong *DD* episodes can reach up to 1 (Fig. ~~S2~~S8-i) while for the extreme episodes (Fig. ~~S2~~S8-ii) it can be as large as 3. The main finding, based on the intercomparison of the two methodologies for the identification of *DD* episodes, is that the frequency of the episodes is higher for the METHOD-B with respect to the primary methodology, while the intensity is decreased. Both facts are expected and can be explained by the lower calculated *AOD* thresholds with METHOD-B thus yielding more *DD* episodes of lower intensity.

4.3 Vertical structure of the Mediterranean desert dust outbreaks

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

Thereby, CALIOP vertical observations were subsequently used to examine the vertical structure of dust outbreaks.

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

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

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4.3.1 Annual characteristics

In Figure 98, are presented the three dimensional structures of the CALIOP overall dust observations (Fig. 98-i) and the associated total backscatter coefficients at 532 nm (Fig. 98-ii), during intense dust episodes conditions, over the broader Mediterranean area, for the period Jun. 2006 – Feb. 2013. From the latitudinal projection in Fig. 98-i, it is evident that dust particles are mainly detected between 0.5 and 6 km, and more rarely up to 8 km, between the parallels 32° N and 38° N. The number of dust observations is increased at higher altitudes with increasing latitudes, up to 40° N, while the altitude range (thickness) where these records are detected is gradually reduced from 4 to 2 km. At northern latitudes, the CALIPSO dust records are drastically reduced and are mainly observed between 1 and 4 km. The ascending mode of the transported mineral particles over the Mediterranean is attributed to the prevailing low pressure systems, which mobilize and uplift dust particles from the source areas across the Sahara Desert and the Arabian Peninsula. Dust aerosols are transported over the planetary boundary layer (Hamonou et al., 1999) due to the upward movement of dry and turbid air masses (Dulac et al., 1992), while the prevailing synoptic conditions determine also the spatial and temporal characteristics of desert dust outbreaks over the Mediterranean (Gkikas et al., 20142015).

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. 98-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., 201~~4~~5) 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~~5-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).

Previous studies have shown that dust layers over the Mediterranean are characterized by a multilayered structure (e.g. Hamonou et al., 1999; Mona et al., 2006; Papayannis et al., 2008). This is also depicted in the longitudinal projection of Figure ~~9~~8-i, where several dust layers of different base/top altitudes and geometrical thicknesses are detected. In general, the base heights vary from 0.5 to 2 km, the top heights from 4 to 6 km and the thicknesses from 1 to 4 km. The majority of common observations between the CALIOP profiles and the identified intense *DD* episodes by the satellite algorithm are recorded over the maritime parts of the study region (bottom map of Fig. 8~~9~~-i). The maximum number of CALIOP dust observations (~ 19000) is recorded along the Atlantic coasts of Morocco, but high numbers (about 10000 – 15000) are also found across the northern African coasts.

Apart from the CALIOP dust observations, we have also analyzed the associated β_{532nm} values at the defined altitude ranges in order to describe the variation of intensity of the desert dust episodes with height over the Mediterranean (Fig. 8~~9~~-ii). The maximum backscatter coefficients (up to 0.006 km^{-1} sr^{-1}) are observed below 2 km, being increased towards the southern edges (30° N - 34° N) of the study region, close to dust source areas.~~The maximum backscatter coefficients (up to 0.006 km^{-1} sr^{-1}) are observed below 2 km, being increased towards the southern edges (30° N - 34° N) of the study region, where their source areas are found~~. This is explained by the fact that dust particles due to their coarse size and large mass, are efficiently deposited and for this reason they are recorded at higher

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concentrations near to the source areas and at low altitudes. Nevertheless, the decreasing intensity with height towards the north is not so evident. Thus, high β_{532nm} values (~ 0.004 km^{-1} sr^{-1}) are observed between 2 and 4 km in the latitudinal zone extending from 35° N to 42° N. Though, the uppermost altitudes where relatively high β_{532nm} values gradually decrease from 6 to 4 km, moving from south to north. Any differences in the latitudinal patterns of dust observations and backscatter values (Figs 98-i and 98-ii) can be explained by the fact that β_{532nm} values take into account only the dust records and not the overall observations (all aerosol types).

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

The longitudinal pattern of β_{532nm} profiles (Fig. 98-ii) is less distinct compared to the corresponding one resulted from the latitudinal projection. Relatively high β_{532nm} values (~ 0.004 km^{-1} sr^{-1}) are found between 1 and 5 km over the western Mediterranean, while over the central and eastern parts of the study region the desert dust outbreaks' intensity (~ 0.006 km^{-1} sr^{-1}) is higher below 1.5 km. Among the sub-regions, the backscatter coefficients are higher in the central and eastern Mediterranean, which is also depicted in the bottom map of Fig. 98-ii. It is reminded that higher intensities of dust episodes over the central and eastern Mediterranean have also been noticed based on MODIS retrievals (Figure 37). From the obtained longitudinal projection, it is evident a patchy structure of the total backscatter coefficient profiles, especially in the central and eastern parts, indicating the existence of several dust layers of varying intensities at different altitudes into the atmosphere.

The three dimensional plots of Figures 8-i and 8-ii, have been also reproduced considering all the available dust and polluted dust CALIOP-CALIPSO records, without taking into account the satellite algorithm's outputs (intense dust outbreaks). The obtained results for the number of observations and β_{532nm} are presented in Figures S98-i and S98-ii, respectively. Note, that for each studied parameter the colorbar scales in Figure 8 and S8 S9 are not identical because the number of observations for dust average conditions (Fig. SS98-i) is extremely larger than the corresponding one during intense dust outbreaks (Fig. 8-i) while the opposite is found for the β_{532nm} values (Fig. 8-ii and Fig. S89-ii). It is apparent that the latitudinal projections calculated for the intense dust outbreaks (Fig. 8-i) and for all the available CALIOP dust records (Fig. S989-i) reveal different patterns. More specifically, when all available CALIOP dust records are considered, it is found that dust aerosols are mainly confined between 1 and 3 km in the southernmost parts of the study region while the number of observations gradually decreases at higher altitudes and towards northern latitudes (Fig. S989-i). On the contrary, during dust outbreaks, mineral particles are transported over the Mediterranean following an ascending path, as it is depicted in the latitudinal projection of Figure 8-i. Nevertheless, it must be mentioned that over the desert areas there is a full coverage (see bottom map in Fig. S989-i) when all dust CALIOP records are considered in contrast to intense dust outbreaks (see bottom map in Fig. 8-i) attributed to the absence of *DT* retrievals, used as inputs to the satellite algorithm, over bright surfaces. The comparison between the longitudinal projections during intense dust outbreaks (Figure 8-i) and during average dust conditions (Fig. S989-i) reveals less remarkable differences than for the latitudinal projections. According to the longitudinal projection of Figure S989-i, in the western Mediterranean, dust layers are confined between 1 and 6 5 km, while their base and top altitude both decrease down to 0.5 and 4.5 km, respectively, for increasing longitudes. In the easternmost part of the study region, dust layers are mainly confined between 1 and 3 km, while its top height can reach up to 5 km. The intensity of dust loads (in terms of β_{532nm}) is lower than 0.003 km^{-1} sr^{-1} regardless the projection plane for average dust conditions based on CALIOP-CALIPSO lidar profiles (Fig. S989-ii). Moreover, the intensity of dust loads decreases gradually with height as well as from south to north revealing a distinct pattern in all projection planes in contrast to the corresponding ones found during desert dust outbreaks (Fig. 8-ii).

4.3.2 Seasonal characteristics

The vertical structure of the Mediterranean desert dust outbreaks has also been analyzed separately for winter (*DJF*), spring (*MAM*), summer (*JJA*) and autumn (*SON*). The seasonal three dimensional representations of the CALIOP overall dust observations and the associated total backscatter coefficients are depicted in the left and right column of Figure 109, respectively. It must be noted, that for β_{532nm} the colorbars' ranges are common, depending on the projection plane. More specifically, the maximum limits have been set to 0.012 km^{-1} sr^{-1}, 0.014 km^{-1} sr^{-1} and 0.021 km^{-1} sr^{-1} for the latitudinal,

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longitudinal and bottom map projections, respectively. It should be mentioned that β_{532nm} values can reach up to 0.045 km^{-1} sr^{-1}, but are associated with a very small number of dust observations.

The majority (85%) of dust observations is recorded in spring and summer, attributed to the enhanced production rates of mineral particles and the prevailing atmospheric circulation over the source areas and the Mediterranean. According to the latitudinal projections, it is evident a seasonal variability of the intense Mediterranean desert dust outbreaks' geometrical characteristics. Dust particles are detected at higher altitudes (6-7 km) during warm seasons of the year while in winter are mainly detected below 3 km and in autumn are recorded between 2 and 5 km. Nevertheless, it should be mentioned that during these seasons only a small number of pixels (see bottom maps in Figs. ~~10~~9 i-a, iv-a) is available considering also that clouds prohibit the satellite observations. Note that in spring, dust can be found at low tropospheric levels while in summer it is mainly observed above 1 km highlighting thus the role of topography and the enhanced thermal convection. During the first half of the year, the maximum dust observations are confined between the parallels 31° N and 37° N while during the second one, are shifted northwards in the latitudinal zone extending from 34° N to 40° N. Similar latitudinal projections were also presented by Luo et al. (2015), for the same zonal areas of the study region, who developed a new algorithm to improve CALIOP's ability to detect optically thin dust layers. From the longitudinal projections as well as from the bottom maps, it is evident that the maximum dust records are found in different Mediterranean sub-regions, depending on the season. The geometrical characteristics, in longitudinal terms, of intense *DD* episodes affecting the western, central and eastern parts of the Mediterranean are similar to those presented in the annual three dimensional structure (Fig. ~~9~~8-i) being more frequent in the eastern and central Mediterranean in winter, spring and autumn and in the western and central Mediterranean in summer.

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The seasonal patterns of β_{532nm} latitudinal projections are different than those for the dust observations, while they also differ among the four seasons. The intensity of winter *DD* episodes is stronger (up to 0.012 km^{-1} sr^{-1}) below 2 km and at the southern parts of the study region. According to the longitudinal and bottom map projections, these episodes take place over the central and eastern Mediterranean Sea but the number of grid cells with coincident CALIOP observations and *DD* episodes is limited. In spring, the highest β_{532nm} values (up to 0.006 km^{-1} sr^{-1}) are recorded between the parallels 31° N and 35° N and below 2 km, although, relatively high β_{532nm} values (up to 0.004 km^{-1} sr^{-1}) are found up to ~~6~~ 5 km (Fig. ~~10~~ 9 ii-b). Moving northwards, over the Mediterranean, dust layers are mainly confined between 2 and 4 km, associated with high β_{532nm} values (up to 0.004 km^{-1} sr^{-1}) in the latitudinal zone extending from 35° N to 43° N. The existence of these elevated dust layers, has been

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also confirmed by model simulations through specific (Papayannis et al., 2008; 2014) or averaged (Alpert et al., 2004) cross sections of dust concentrations in the central sector of the Mediterranean. This is in accordance with our longitudinal projection (Fig. 10 9 ii-b), where β_{532nm} is high varying from 0.004 to 0.008 km^{-1} sr^{-1} at these altitude ranges.

In summer, the intensity of dust episodes is smoothly decreased at higher altitudes, where dust layers of considerable β_{532nm} values are also found. More specifically, the highest backscatter coefficients (up to 0.008 km^{-1} sr^{-1}) are recorded near to the surface but also moderate values (up to 0.006 km^{-1} sr^{-1}) are observed between 2 and 5 km, particularly over the southern parts of the study region (Fig. 109 iii-b). Most of these intense *DD* episodes occur in the western Mediterranean, where the highest β_{532nm} values (up to 0.005 km^{-1} sr^{-1}) are recorded between 2 and 5 km. Over the central and eastern Mediterranean, even higher β_{532nm} values are found (up to 0.014 km^{-1} sr^{-1}) but at lower altitudes (< 1 km). In autumn, the majority of the grid cells of coincident CALIOP profiles and *DD* episodes identified by the satellite algorithm are located between the parallels 33° N and 41° N. In this latitudinal zone, CALIOP profiles are available over the interior parts of the Iberian Peninsula and over western and central parts of the Mediterranean Sea, near to the northern African coasts. According to the latitudinal projection, β_{532nm} values mainly vary from 0.002 to 0.009 km^{-1} sr^{-1}, revealing an increasing tendency for increasing heights. On the contrary, the total backscatter coefficients do not show a distinct spatial pattern on the longitudinal projection, due to the limited number of grid cells participating in the calculations. Throughout the year, based on the CALIOP β_{532nm} retrievals, the *DD* episodes are more intense (up to 0.018 km^{-1} sr^{-1}) in spring, when massive dust loads are transported from the Sahara desert towards the central and eastern parts of the Mediterranean Sea (bottom map in Fig. 10 9 ii-b).

4.4. Intercomparison of satellite AOD and PM_{10} concentrations for specific desert dust outbreaks

In Section 4.21.2, it has been shown that the agreement between the satellite algorithm's outputs and PM_{10} concentrations is better in the central and eastern Mediterranean with regards to the western parts (Figure 85-ii). This discrepancy has been mainly attributed to the higher altitude of dust layers' base over the western sector of the study domain (Figure 98-i), in relation to the existing areal orography. Here, aiming at addressing how dust layers' geometrical characteristics influence the agreement between columnar *AOD* satellite and ground PM_{10} measurements, specific desert dust outbreaks that took place over the PM_{10} stations are analyzed. These outbreaks were selected based on

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concurrent fulfillment of the following criteria: (i) a *DD* episode must be identified by the satellite algorithm at pixel level (at 1° x 1° grid cell), (ii) total PM_{10} measurement must be available at the station which lies into the geographical limits of the corresponding grid cell and (iii) CALIPSO flies across the grid cell. These criteria were met for 13 desert dust outbreaks, which took place over 9 PM_{10} stations during the period ~~2000-2013~~Jun. 2006 – Feb. 2013. Similarities were found among the identified cases and therefore only the results for four desert dust outbreaks of different geometrical characteristics are discussed in the present section. For each case, we have reproduced the cross sections of the β_{532nm} vertical profiles up to 8 km above sea level (a.s.l.) along the CALIOP-CALIPSO track when the satellite flies near the PM_{10} site (Figures 1~~1~~0-1~~3~~2). Moreover, the corresponding aerosol subtype profiles, acquired from the CALIOP website (http://www-calipso.larc.nasa.gov/products/lidar/browse_images/production/), are provided in the supplementary material (Figures S10~~S7~~9-S~~9~~1~~1~~12). Since the PM_{10} concentrations are available only as daily averages, the optimum solution would be to have the maximum number (2) of CALIOP overpasses near PM_{10} site throughout the day, in order to reduce the temporal inconsistencies between satellite vertical resolved retrievals and ground data. However, in 8 out of 13 desert dust outbreaks this was not feasible.

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

The first study case refers to a desert dust outbreak that took place on 26th May 2008 and affected the station Censt (Lat: 39.064, Lon: 8.457) located in southern Sardinia. At the ground, the measured mean daily total PM_{10} concentration was 19 μg m^{-3} whereas 68% (or 13 μg m^{-3}) of the load consisted of dust particles indicating thus their strong presence in the lowest troposphere. Based on MODIS-Terra retrievals, representative for the whole atmospheric column and grid cell, the aerosol optical depth at 550 nm was equal to 0.81. In order to investigate the vertical distribution of the dust outbreak, the cross sections of the β_{532nm} vertical profiles along CALIOP track, near the station, during daytime and nighttime have been reproduced and depicted in Figures 1~~1~~0-i and 1~~1~~0-ii, respectively. In addition, the corresponding aerosol subtype profiles are provided in Figures S~~7~~~~10~~109-i and S~~7~~~~10~~109-ii in the supplementary material. During night, it is evident the predominance of a well-developed dust layer mixed with polluted aerosols (Figure S~~7~~109-i) extending from surface up to 5 km a.s.l. between the parallels 33° N and 38° N, while near the station its top is lowered down to 3 km (left side of Figure 1~~1~~0-i). Moreover, the β_{532nm} values range mainly from 0.002 to 0.003 km^{-1} sr^{-1} without revealing remarkable variations, thus indicating a rather compact dust layer. According to the daytime CALIOP overpass (Figure 1~~1~~0-ii), a pure dust layer (Figure S~~7~~10-ii) is confined between surface and 4 km, affecting the surrounding area of the station, while its intensity (in terms of β_{532nm}) varies slightly from 0.0015 to 0.002 km^{-1} sr^{-1}. Nevertheless, due to the background solar illumination, leading thus to a lower signal-to-noise ratio (Nowottnick et al., 2015), the "borders" of the dust plume during daytime are not so distinct in contrast to nighttime. According to the obtained results, the ground-based measurements are able to capture satisfactorily the dust event when its load is equally distributed in the lowest tropospheric levels, resulting thus to a good agreement between MODIS and PM_{10} observations.

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

Two dust events that affected Els Torms (NE Spain, Lat: 41.395, Lon: 0.721) and San Pablo (central Spain, Lat: 39.525, Lon: -4.353) on 16th July 2008 and 12th September 2007, respectively, are studied here. The daily averages of the total PM_{10} concentrations were equal to 16 and 30 μg m^{-3}, respectively, whereas the dust particles' contribution (dust PM_{10}) to the total amount was zero in Els Torms and 33 % in San Pablo. On the contrary, the MODIS-Terra level 3 *AOD* retrievals were high and equal to 0.56 (Els Torms) and 0.64 (San Pablo), indicating the existence of dust aerosols according to the satellite algorithm's classification method. In order to give a better insight, aiming at describing the discrepancies between MODIS-Terra *AOD* and PM_{10} concentrations, we have reproduced the cross sections of the total backscatter at 532 nm when CALIPSO flies, during daytime, near Els Torms (Figure 1~~2~~1~~1~~-i) and San Pablo (Figure 1~~2~~1~~1~~-ii). The corresponding profiles of the CALIOP aerosol classification scheme are also available in Figures S~~8~~~~11~~1~~0~~1-i and S~~8~~~~11~~1~~0~~1-ii. In Els Torms, where the dust PM_{10} concentration was zero, a dust layer (Figure S~~8~~~~11~~1~~0~~1-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 1~~2~~1-i). Through CALIOP lidar profiles, it is confirmed the existence of a dust layer aloft, which cannot be captured by the PM_{10} measurements in contrast to the MODIS spectroradiometer. In San Pablo, where the dust particles' contribution to the total PM_{10} load was equal to 33 %, a dust layer abuts the ground extending up to 5-6 km ~~ASL~~a.s.l., whereas the dust plume covers a wide range, in latitudinal terms, from the sub-Sahel to the Celtic Sea, affecting the Iberian Peninsula (Figure S~~8~~11-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 11~~2~~-ii. The two studied cases here differ from

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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 PM_{10} 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 PM_{10} concentration was equal to 134 μg m^{-3} accounting for the 92 % of the total PM_{10} 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,~~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 PM_{10} site (Agia Marina) extends from close to the ground up to 9 km a.s.l., comprising only pure dust aerosols (Figure S~~91~~12). 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. where the maximum β_{532nm} values (up to 0.006 km^{-1} sr^{-1}) are observed (Figure 1~~3~~2). Also, similar β_{532nm} values are recorded below 1 km a.s.l.; however, the dust layer is not well represented in the cross section of the CALIOP β_{532nm} vertical profiles due to the total attenuation of the lidar beam by clouds (located between 3 and 4 km a.s.l.) superimposed to the low-elevated dust layer.

5. Summary and conclusions

This study aims at describing the vertical structure of intense desert dust outbreaks affecting the broader Mediterranean basin. To achieve this target, an updated version of an objective and dynamic algorithm, which has been introduced by Gkikas et al. (2009; 2013), has been applied for the identification of strong and extreme desert dust episodes, over the period Mar. 2000 – Feb. 2013. For its operation, a group of optical properties, retrieved by satellite sensors (MODIS-Terra/Aqua, EP-TOMS and OMI-Aura) on a daily basis, is used, providing information about aerosols' load, size and nature. Briefly, the satellite algorithm consists of three parts; at the first one are computed the mean *AOD* value (*Mean*) and the associated standard deviation (*Std*) for the whole study period in each grid

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cell of 1° x 1° spatial resolution, at the second one the identified aerosol episodes are classified based on their intensity to strong and extreme ones. Finally, at the third part the identified aerosol episodes are categorized as desert dust episodes, separately over land and sea. Through this approach the selected dataset consists only of intense desert dust episodes since their intensity (expressed in terms of AOD_{550nm}) is higher/equal than *Mean + 2*Std*. The *DD* episodes have also been determined by applying an alternative second methodology (METHOD-B) which excludes dust-affected cases identified based on the criteria set concerning the aerosol size/nature related optical properties.

Through the comparison of the default version of the satellite algorithm against surface measurements derived from 109 AERONET and 22 PM_{10} 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 (~ 0.25 μm^3 μm^{-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 $\alpha_{440-870nm}$ 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.

PM_{10} and dust contribution

- The agreement between surface and satellite measurements is better over the central and eastern Mediterranean stations.
- On a station level, the percentage of the intense *DD* episodes, for which a dust contribution to PM_{10} surface concentration has been recorded, varies from 68% (Monagrega, northeastern Spain) to 97% (Boccadifalco, Sicily).
- In the majority of stations, dust particles contribute more than 50% of the total amount reaching up to 86.8% (Agia Marina, Cyprus).

- The mean PM_{10} concentration levels mainly vary from 20 to 50 μg m^{-3} reaching up to 223 μg m^{-3} in Agia Marina (Cyprus).

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

- Strong *DD* episodes occur more frequently (up to 9.9 episodes yr^{-1}) in the western Mediterranean while the extreme ones occur more frequently (up to 3.3 episodes yr^{-1}) over the central parts of the Mediterranean Sea, when the satellite algorithm operates with MODIS-Terra 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.

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

- Dust particles are mainly detected between 0.5 and 6 km, following an ascending mode, up to 40º N, leaving from the source areas and transported towards the Mediterranean.

- Over the western Mediterranean, the dust layers are mainly observed between 2 and 6 km while their base height is decreased down to 0.5 km for increasing longitudes.
- During the warm period of the year, dust particles are uplifted at higher altitudes (up to 8 km).
- In summer, the transported dust loads over the western Mediterranean are recorded above 1 km and in spring at lower altitudes over the central and eastern parts of the study region. This behavior underlies the role of topography (e.g. Atlas Mountains) and the enhanced thermal convection.
- The intensity of dust outbreaks, in terms of β_{532nm}, is maximized (up to 0.006 km^{-1} sr^{-1}) below 2 km and at the southern parts (30° N - 34° N) of the study region.
- In spring, considerably high β_{532nm} values (~ 0.004 km^{-1} sr^{-1}) are observed between 2 and 4 km in the latitudinal zone extending from 35° N to 42° N.
- Moderate-to-high β_{532nm} values are observed up to 6 km, near to the source areas, while the top of dust layers is gradually decreased down to 4 km towards northern latitudes.
- From the longitudinal projection of β_{532nm}, it is evident that *DD* episodes are more intense (~ 0.004 km^{-1} sr^{-1}) between 1 and 5 km in the western Mediterranean, while over the central and 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 km^{-1} 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~~ Italy (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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Table 1: AERONET stations, depicted with cyan colors in Figure ~~4~~1, which have been used for the identification of desert dust (*DD*) episodes based on ground retrievals.

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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

Table 2: Percentages of the satellite Ångström exponent, Fine fraction, Effective Radius and Aerosol Index retrievals 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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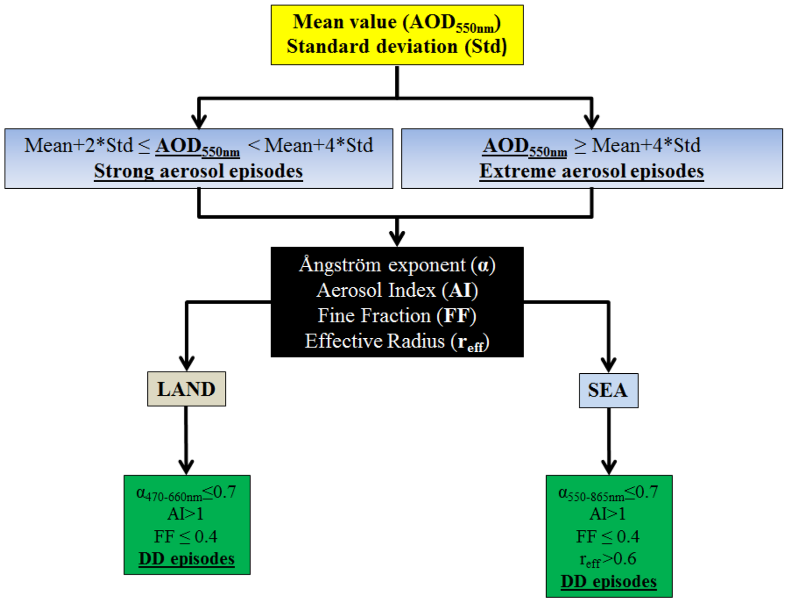

~~**Figure 1:** Methodology applied to each 1° x 1° grid cell for the identification of the intense Mediterranean desert dust outbreaks.~~

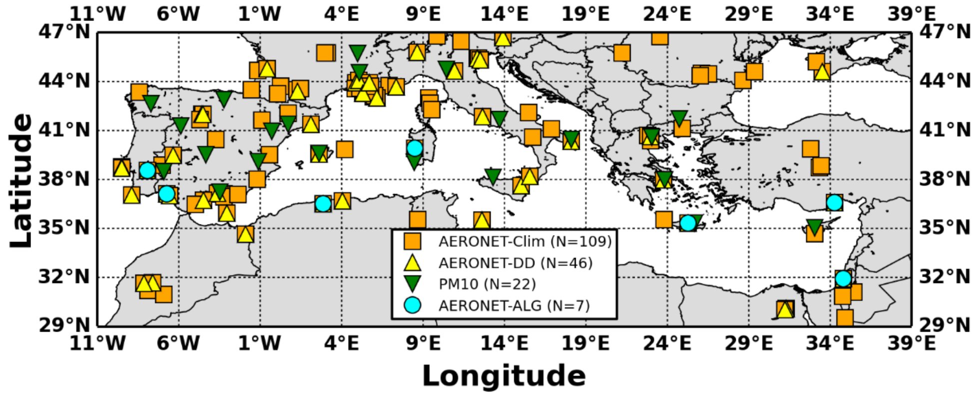

Figure 41: Locations of the AERONET and PM_{10} stations which have been used for the evaluation of the algorithm's outputs. More specifically, with orange squares are denoted the AERONET stations located into the study region, with the yellow triangles the AERONET stations with coincident satellite 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 PM_{10} stations.

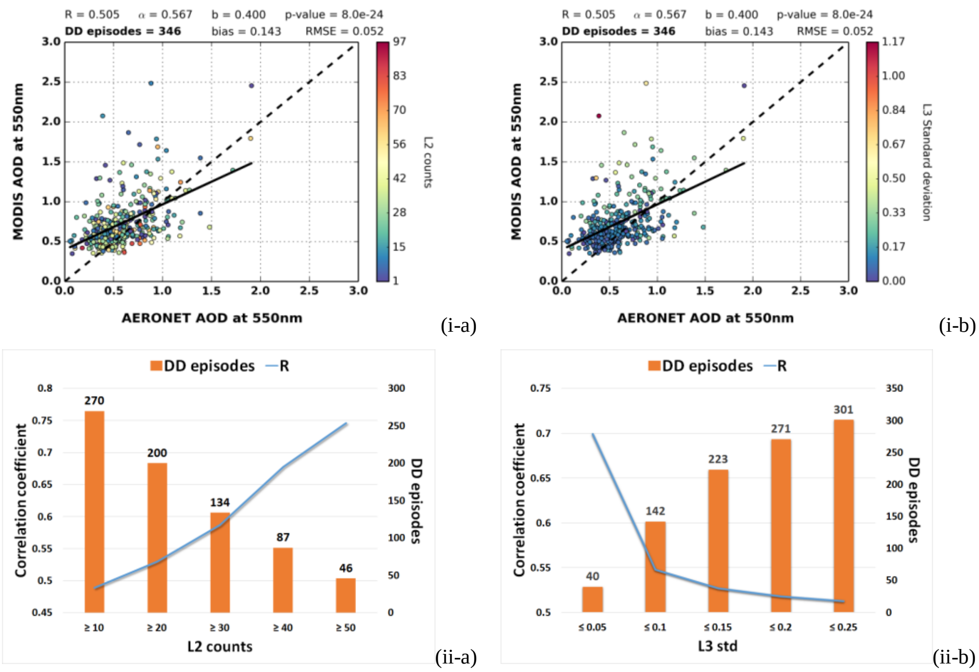

Figure ~~5~~2: (i) Scatterplots between MODIS-Terra and AERONET aerosol optical depths at 550 nm under intense desert dust episodes conditions related to the: (a) number of level 2 counts which are used for the calculation of the level 3 retrievals and (b) spatial standard deviation inside the 1° x 1° 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.

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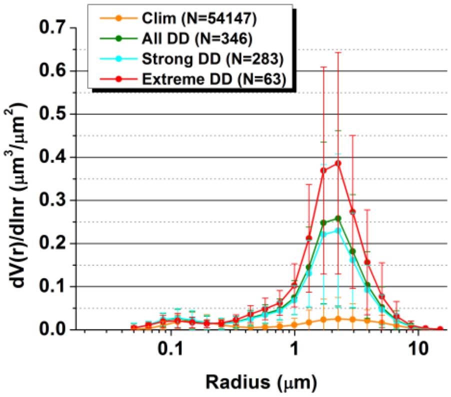

Figure 36: AERONET size distributions averaged for all available retrievals (orange curve) as well as for the total (green curve), strong (cyan curve) and extreme (red curve) desert dust episodes, occurred over the broader area of the Mediterranean basin, during the period Mar. 2000 – Feb. 2013. The error bars represent the calculated standard deviations.

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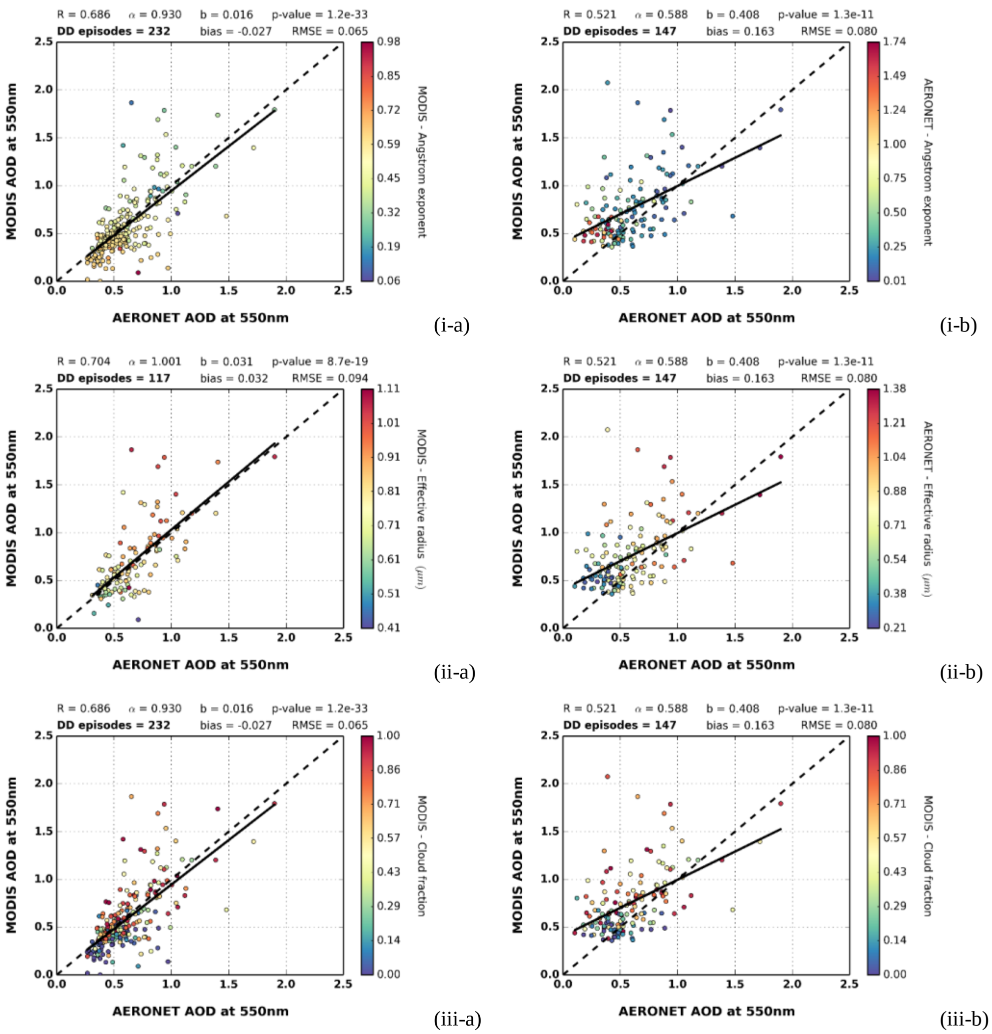

Figure ~~7~~4: Scatterplots of MODIS-Terra and AERONET aerosol optical depths at 550 nm when intense dust episodes have been identified based on: (a) AERONET retrievals and (b) satellite algorithm, respectively. In the left column, colormaps indicate the corresponding values of: (i) Ångström exponent, (ii) Effective radius and (iii) Day cloud fraction derived by MODIS-Terra retrievals. In the right column, colormaps indicate the corresponding values of: (i) AERONET Ångström exponent, (ii) AERONET Effective radius and (iii) MODIS day cloud fraction retrievals. For each scatterplot, are provided the correlation coefficient (*R*), slope (*α*), intercept (*b*), p-value, number of *DD* episodes, bias (MODIS – AERONET) and root mean square error (*RMSE*).

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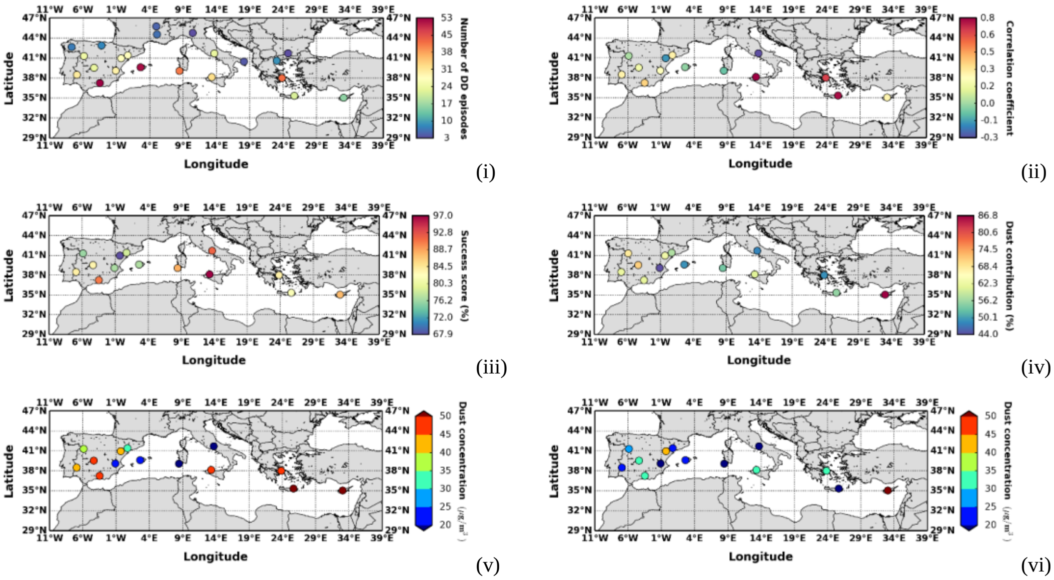

Figure 58: (i) Number of concurrent intense *DD* episodes where total PM_{10} concentrations and MODIS-Terra *AOD* retrievals are available, (ii) Computed correlation coefficient values between total PM_{10} concentrations and MODIS-Terra *AOD* retrievals in stations where at least 10 *DD* episodes have 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 (μg m^{-3}), based on ground measurements for the identified intense *DD* episodes by the satellite algorithm.

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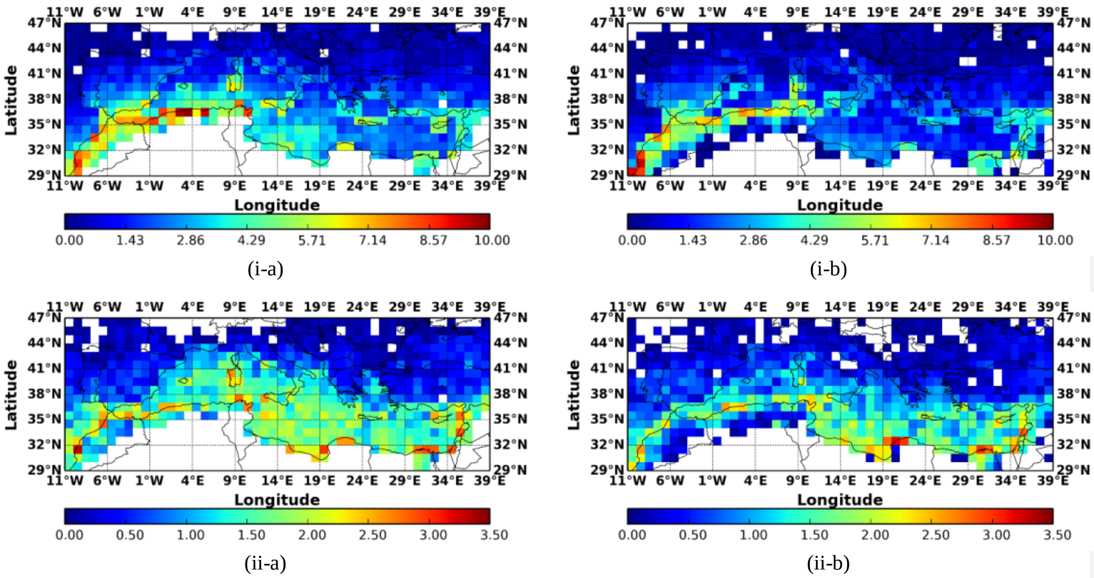

Figure 26: Geographical distributions of the occurrence frequency (episodes/ yearyr^{-1}) of: (i) strong and (ii) extreme desert dust episodes, averaged for the periods: (a) Mar. 2000 – Feb. 2013 (MODIS-Terra) and (b) 2003 – 2012 (MODIS-Aqua), over the broader area of the Mediterranean basin.

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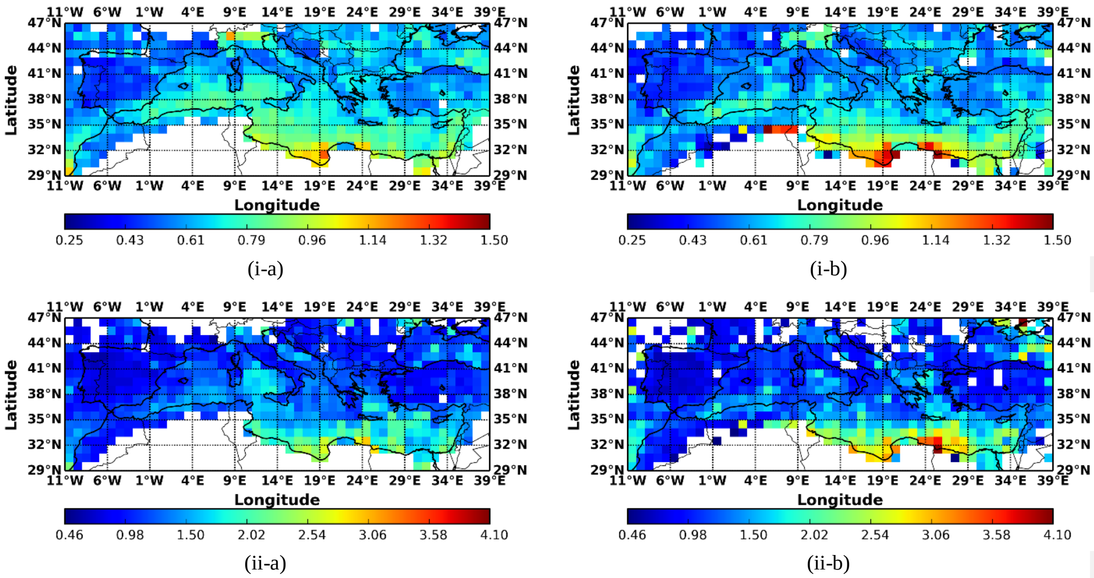

Figure 37: Geographical distributions of the intensity (in terms of AOD_{550nm}) of: (i) strong and (ii) extreme desert dust episodes, averaged for the periods: (a) Mar. 2000 – Feb. 2013 (MODIS-Terra) and (b) 2003 – 2012 (MODIS-Aqua), over the broader area of the Mediterranean basin.~~(a) 2000 – 2013 and (b) 2003 – 2012, over the broader area of the Mediterranean basin.~~

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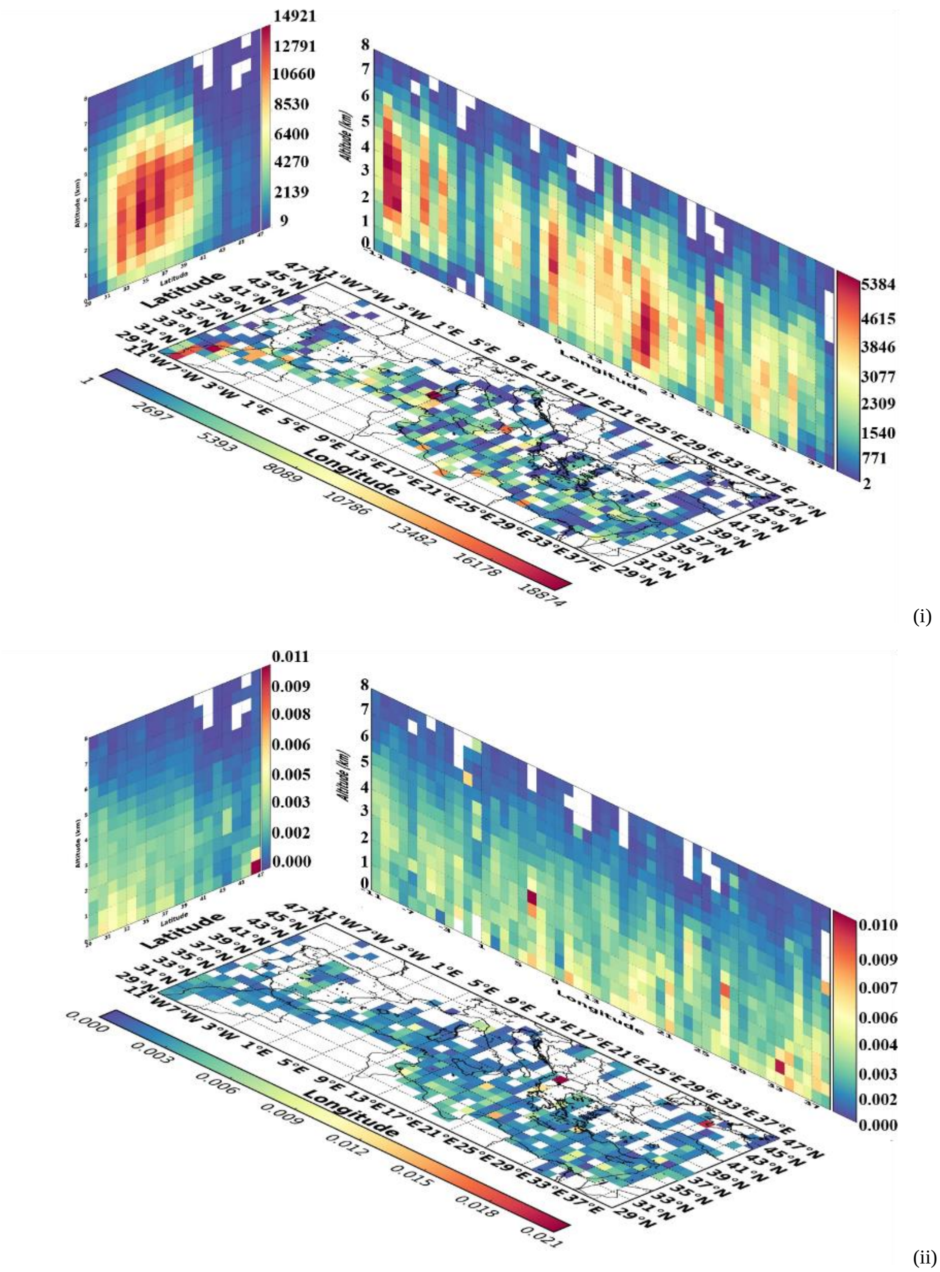

Figure 89: Three dimensional structure of the: (i) overall number of dust and polluted dust observations and (ii) total backscatter coefficient at 532 nm (in km^{-1} sr^{-1}), over the broader Mediterranean basin under *DD* episodes conditions, based on CALIOP-CALIPSO vertical resolved retrievals for the period Jun. 2006 – Feb. 2013.

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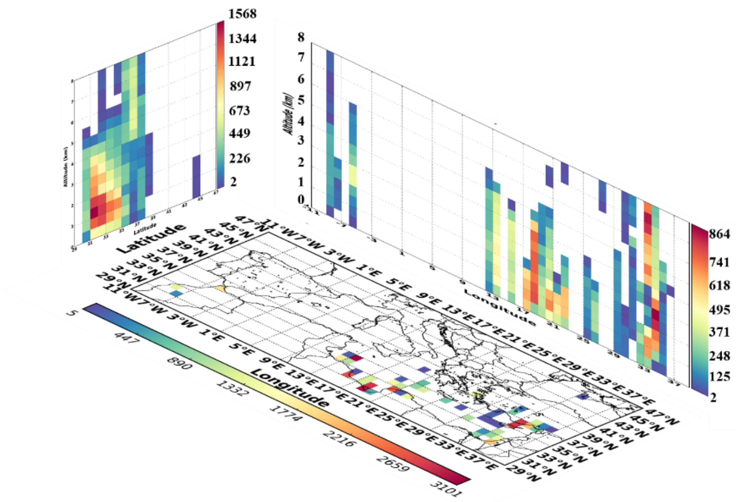
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Latitude
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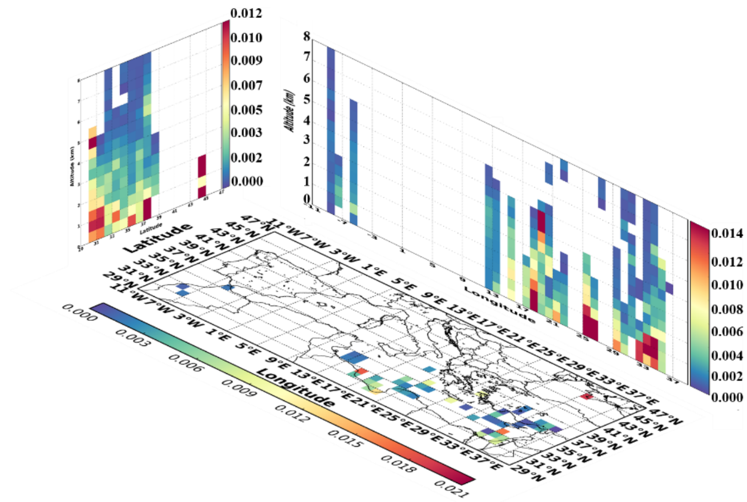
(i-a)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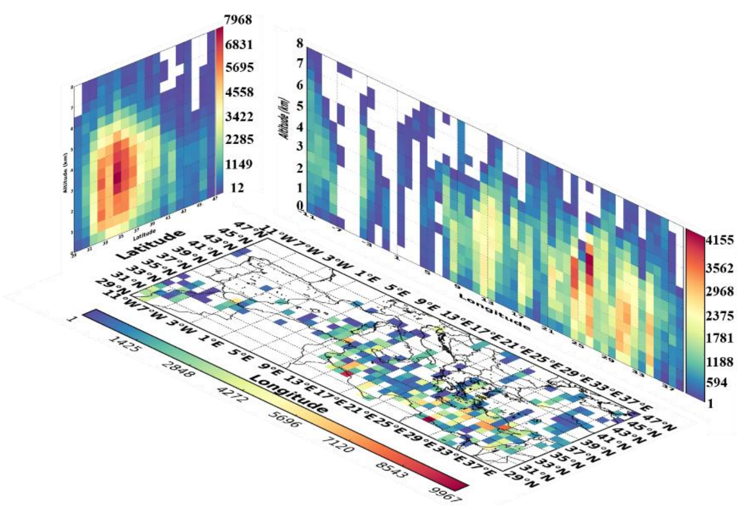
(i-b)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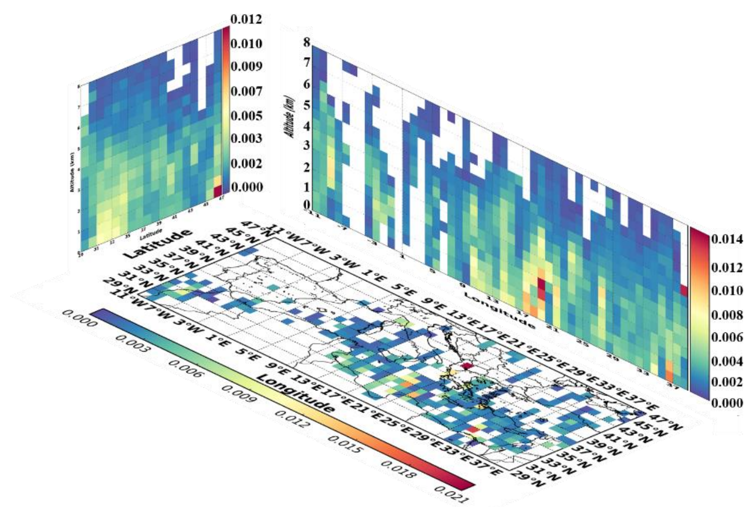
(ii-a)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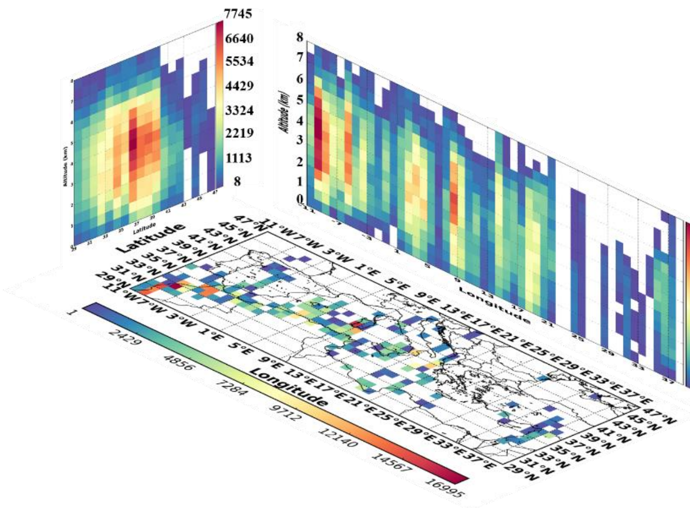
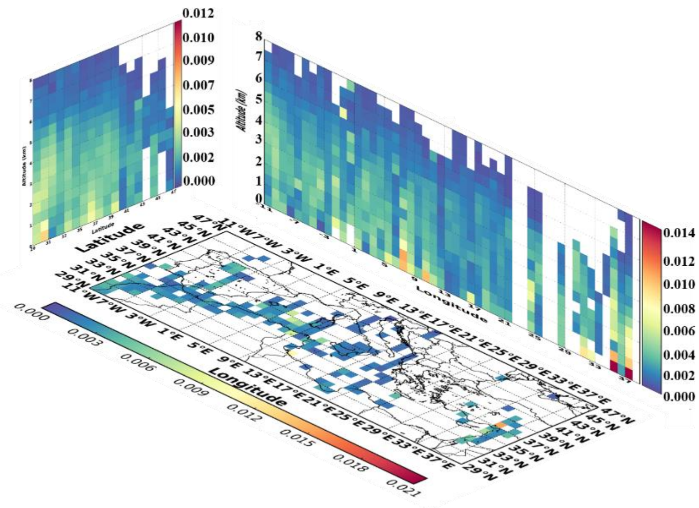
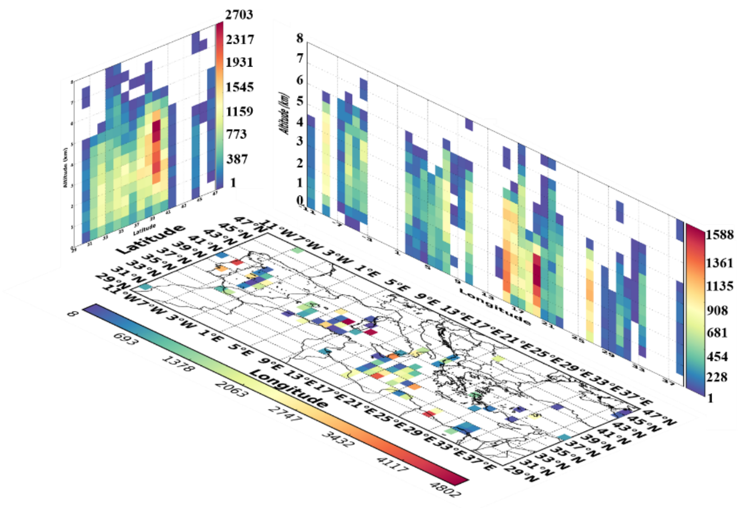
(ii-b)



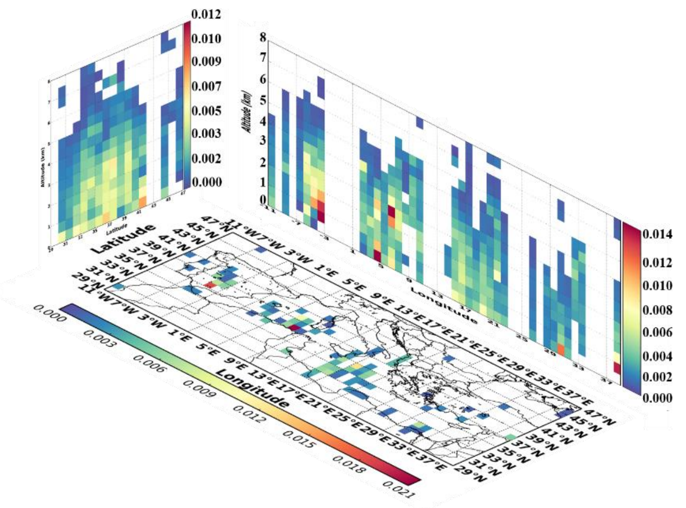
(iii-a)



(iii-b)



(iv-a)



(iv-b)

Figure ~~9~~10: Three dimensional representation of the: (a) overall number of dust and polluted dust observations and (b) total backscatter coefficient at 532 nm (in km^{-1} 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.

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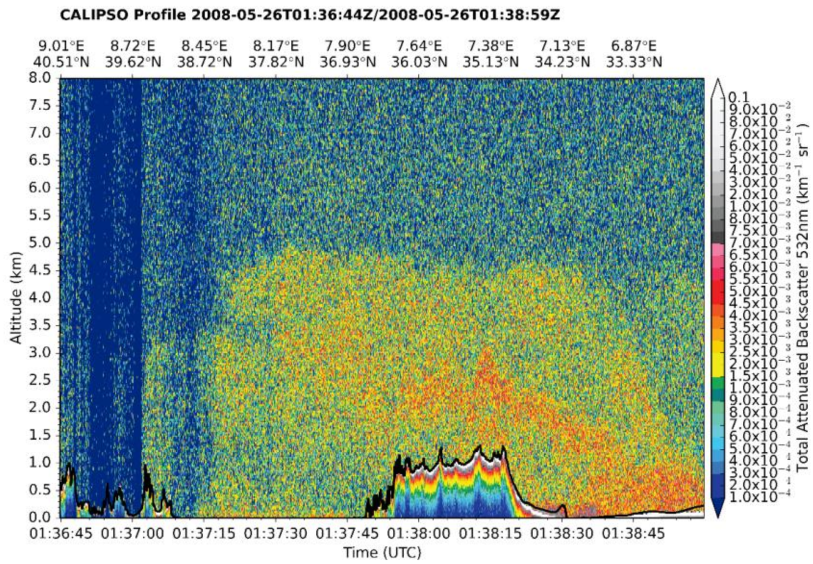
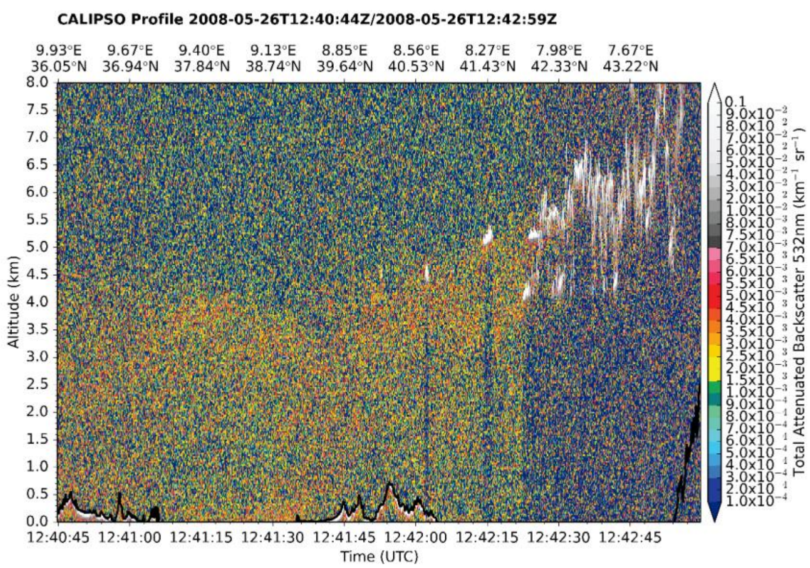


(i)



(ii)

Figure 10~~1~~: Cross sections of the total backscatter coefficient at 532 nm (in km^{-1} 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.

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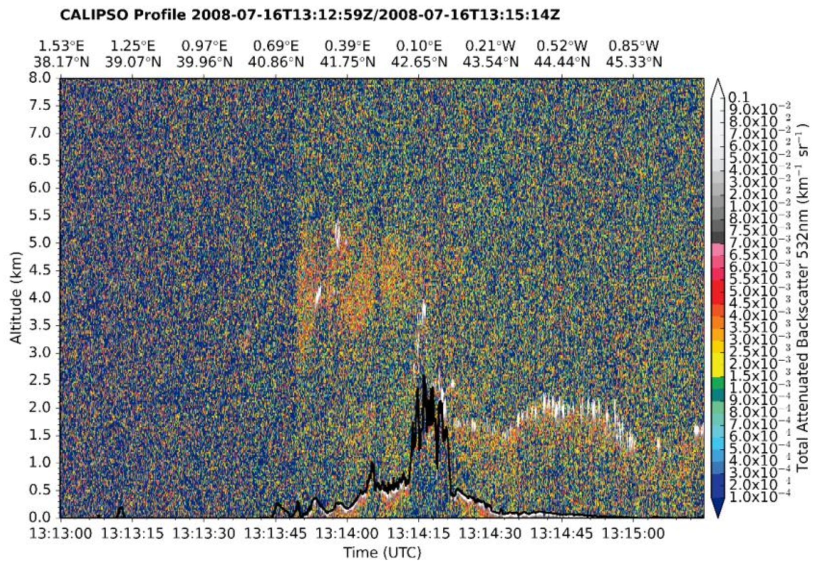
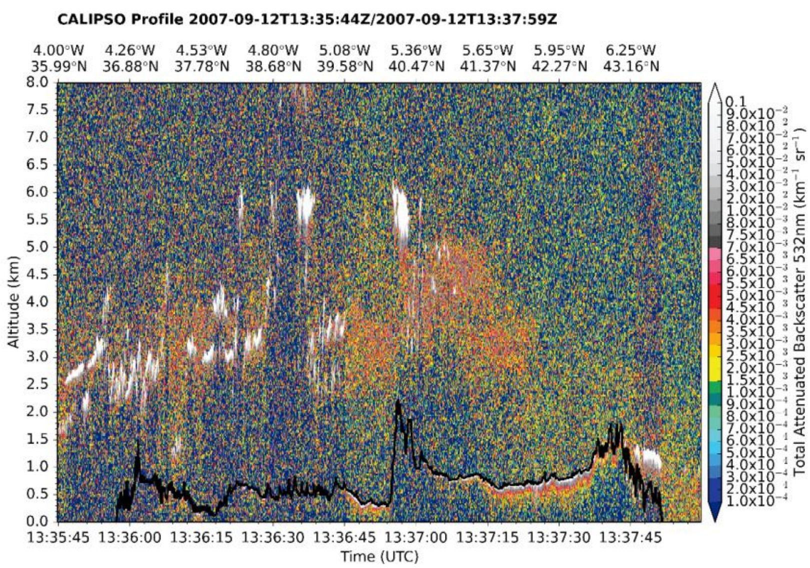


(i)



(ii)

Figure 112: Cross sections of the total backscatter coefficient at 532 nm (in km^{-1} 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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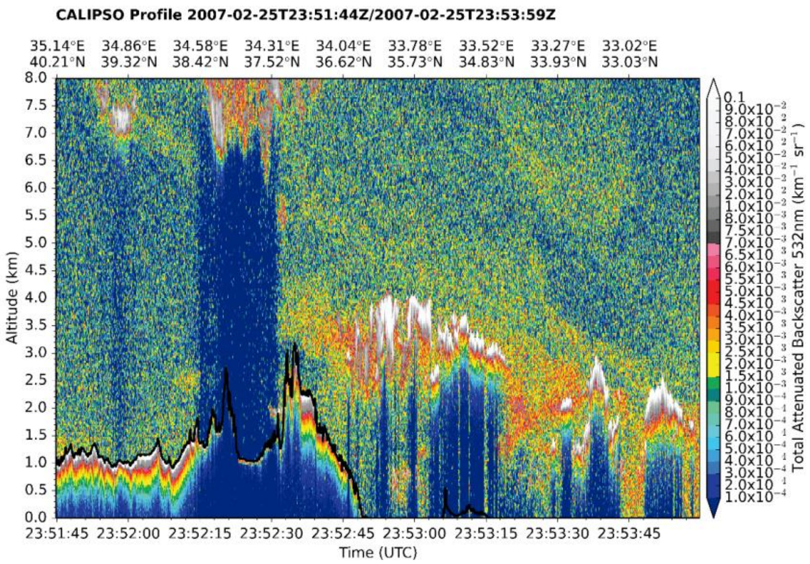

Figure 12~~3~~: Cross section of the total backscatter coefficient at 532 nm (in km^{-1} sr^{-1}) vertical profiles along the CALIOP-CALIPSO track during ~~daytime~~ nighttime over the station Agia Marina (Lat: 35.039, Lon: 33.058) on 25th February 2007. The black thick solid line represents the surface elevation.

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