Response to Reviewers

We would like to thank the Reviewer for the comments which helped us to prepare a revised version of our manuscript. We have taken into account these suggestions and addressed the raised issues. Below are given point by point answers to the comments (in Italics).

Reviewer 3

However, two main questions arise, which are not completely ignored by the authors, but seriously weaken the scientific relevance of this work. First, all the input data are integrated (aerosol optical depth) or averaged (aerosol single scattering albedo and asymmetry parameter) over the whole atmospheric column, while it is well known that aerosol vertical profiles show strong gradients. At least a sensitivity test should have been performed to determine how the various aerosol direct radiative effects (DREs) discussed in the manuscript are sensitive to changes in the aerosol vertical distribution.

Indeed, as mentioned by the Reviewer, reference to the issue of vertically resolved aerosol properties was made in the ACPD paper, more specifically in the last paragraph of the Conclusions. There we explained why we did not account for vertical distributions of aerosol properties in the present study and mentioned that this will be the focus of future work.

However, a sensitivity test can help to assess the uncertainties that are introduced by the use of columnar instead of vertically changing aerosol properties in our radiative transfer model (RTM). Therefore, following the Reviewer's suggestion, we have performed a series of model sensitivity tests, in which the RTM input aerosol properties changed with height. Here, it should be noted that among the three key model aerosol optical properties, i.e. aerosol optical depth (AOD), single scattering albedo (ω_{aer}) and asymmetry parameter (g_{aer}), relevant and homogeneous vertically resolved information over the Mediterranean basin is available only for the first one, i.e. AOD, from recent satellite lidar measurements (CALIOP/CALIPSO). Limited information on changing aerosol ω_{aer} with height is available from sporadic measurements at specific Mediterranean sites, whereas no relevant information is available on g_{aer}. Moreover, it should be noted that, according to the existing literature, aerosol loading (mass) is the aerosol property that changes more drastically with height than others. Thus as a first step, we have tried to assess the uncertainty of model computed aerosol direct radiative effects (DREs), namely at the top of atmosphere (TOA, DRE_{TOA}), in the atmosphere (DRE_{atm}) and at the surface (DRE_{netsurf}), to changing AOD with height. The sensitivities were performed as follows.

We run the RTM using: (i) columnar integrated AOD values (as done in the original paper), and (ii) AOD vertical profiles, i.e. AOD values for 40 layers in the atmosphere extending from the surface, constrained by topography, up to the top of atmosphere. The AOD data were taken from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP Level 2-Version 3.01) instrument onboard the CALIPSO satellite (launched in April 2006, Winker et al., 2007). The RTM was run for 4 geographical cells, representative of different aerosol types prevailing over the Mediterranean basin. The four cells, whose altitudes range from 90m to 350m, are centered at:

- (a) 38.75° N, 23.75° E, including Athens (Greece), corresponding to urban environment
- (b) 31.25° N, 6.25° E, in Algeria (north Africa), corresponding to desert environment
- (c) 38.75° N, 8.75° E, in the western Mediterranean Sea, corresponding to marine environment
- (d) 46.25° N, 21.25° E, in the Romanian plains, corresponding to continental environment

The vertical profiles of AOD, based on CALIOP data, are shown in Figure 1. The columnar integrated values of AOD are also given in the same Figure. It should be noted that the large columnar AOD value for the maritime environment, along with the existence of aerosols at altitudes 6-7 Km, and 8-10 Km, indicate that this is a case in which probably desert dust co-exists with maritime aerosol, which is present in the lower troposphere. This is not strange given the location of the marine geographical cell, in the Mediterranean Sea between Algeria and Corsica.



Figure 1. Vertical profiles of AOD, based on CALIOP data, over four Mediterranean geographical cells characterized by urban, dust, maritime and continental aerosols. Columnar integrated AOD values are also given.

The runs of RTM were performed for July 2007. This date (month) was chosen in order to ensure overlapping between the CALIOP AOD availability and our study period, and also an upper bound limit of uncertainties, given that, as shown in our study (e.g. Fig. 5), DRE values peak in summer.

The differences (in W m⁻²) between the model computed aerosol DREs using columnar and vertically resolved aerosol information are given in Figure 2, separately for each one of the four different aerosol types, and for every aerosol DRE component (DRE_{TOA} , DRE_{surf} and $DRE_{netsurf}$).

Inclusion of the vertical distribution of AOD in the RTM modifies aerosol DREs by amounts ranging from about -2 to 2 W m⁻², depending on the aerosol type. In general, the largest differences are found for marine and continental aerosols. In particular, in terms of DRE_{TOA}, continental aerosol is found to be most sensitive to the vertical variation of AOD, with a change equal to 1.9 W m⁻², whereas at the surface (DRE_{surf} and DRE_{netsurf}) the largest sensitivity is found for maritime aerosol, with DRE changes equal to 0.4 W m⁻². The percent differences, not shown here, vary between -5 and 3%.

In conclusion, using columnar instead of vertically distributed AOD values in the RTM, introduces relatively small uncertainties in the computed aerosol direct radiative effects, with magnitudes smaller than 2 W m^{-2} or 5%, in absolute and relative percentage terms.



Figure 2. Absolute differences (in $W m^{-2}$) between model DRE components at TOA, in the atmosphere and at the surface, computed using columnar and vertically resolved AOD over four Mediterranean geographical cells characterized by urban, dust, maritime and continental aerosols.

We have further investigated the potential sensitivity of aerosol DREs to changing ω_{aer} with height. In these tests, we relied on the same conditions ruling the sensitivities of AOD described before. Hence, we have considered two cases in which dust aerosols were overlying (at altitudes from 1 to 5 km) a boundary layer including urban and maritime aerosols. The differences (in W m⁻²) between the model computed aerosol DREs using changing ω_{aer} values with height, i.e. including two different aerosol types (urban-dust, and maritime-dust), and constant ω_{aer} values with height, i.e. single aerosol types (urban and maritime), are given in Figure 3, separately for the two cases, and for every aerosol DRE component (DRE_{TOA}, DRE_{surf} and DRE_{netsurf}). It is seen again that taking into account the vertical distribution of ω_{aer} in the RTM modifies aerosol DREs by amounts ranging from about -2 to 2.5 W m⁻², depending on the aerosol component and type.



Figure 3. Absolute differences (in W m^{-2}) between model DRE components at TOA, in the atmosphere and at the surface, computed using constant and changing ω_{aer} with height over two Mediterranean geographical cells characterized by urban and maritime aerosols in the boundary layer. Dust aerosols are overlying the boundary layer in the cases of changing ω_{aer} .

We did not include the specific Figures in the manuscript as the paper is already lengthy enough. Thus, we have provided a short discussion in the last paragraph of Conclusions (section 5), which is now renamed to "Discussion and Conclusions .

Second, the aerosol optical properties (which are retrieved from passive remote sensing instruments) are affected by random and possibly systematic errors, which may be difficult to assess although they are perhaps as large as the errors of the aerosol optical properties calculated by global models. Both these aspects could well hamper the work by Papadimas and co-authors to reduce the current uncertainty of the aerosol direct forcing over the Mediterranean compared to the current knowledge.

Both satellite and modeling results require evaluation against higher quality observational products derived from surface or in-situ measurements. MODIS AOD data over the Mediterranean basin have been extensively and successfully validated against AERONET station data (e.g. Papadimas et al., 2008 and 2009, among others) and that such satellite data provide the complete spatial and temporal variability that is not possible from ground-based measurements. Furthermore, the present study provides fine spectral resolution computations of solar fluxes and aerosol radiative effects, and thus of high accuracy, which are obtained with a detailed spectral radiative transfer model. Therefore the obtained DREs (and fluxes) ensure a much finer spectral resolution than those obtained by global models that usually consider a few broad spectral bands, which has been shown (e.g. Hatzianastassiou et al., 2004b, 2007) to critically affect DREs. The present work provides, for the first time to our knowledge at least at the spatial and temporal scales and coverage of this study, computations of aerosol DREs under realistic surface and atmospheric conditions. The present study combines MODIS aerosol information with a detailed spectral radiative transfer model, to ensure DRE computations of quality over the Mediterranean basin.

Yet, this manuscript would have been useful if it had clearly highlighted the role of various variables and parameters in the spatial and temporal variations of the aerosol radiative forcing (as suggested in the abstract).

The present study's aim is to compute the spatial and temporal variability of DREs in the Mediterranean basin and to analyze the DRE dependence on the key radiative properties of aerosols, namely AOD, ω_{aer} and g_{aer} .

What is proposed by the reviewer is far beyond the scope of the present, already sufficiently comprehensive study, and the determination of the role of the various parameters is an important issue that certainly deserves to be studied in a future work. In such a work, however, further analysis should be undertaken, including also non-aerosol properties, which also affect DREs, namely, cloud cover, incoming solar radiation or surface albedo.

But the discussions regarding this question are too often obscure and not convincing because they are not straightforward enough. For instance, sections 3.2 and 3.3 compare DRE ratios to AOD ratios for various cases, while the comparison of the aerosol radiative efficiencies (E)would provide with the same information in a smarter way. Instead, section 3.4 discusses E as a possible mans to estimate aerosol forcing which is little rigorous.

As clarified above, we have not attempted to analyze the role of various parameters on aerosol DREs. In sections 3.2 and 3.3 the patterns of spatial and temporal variability of the various components of aerosol DREs, i.e. DRE_{TOA} , DRE_{atm} , and $DRE_{netsurf}$, are discussed, whereas the role of scattering and absorption abilities of aerosols for DREs, by means of the ratio $DRE_{TOA}/DRE_{netsurf}$, is also examined in section 3.3. Furthermore, the aerosol radiative

efficiency, E_{AOD} , is examined and discussed in section 3.4. It should be noted that E_{AOD} does not provide the same information with the aforementioned ratios, since it quantifies the dependence of aerosol DREs on AOD, solely, i.e. the perturbation of solar radiative fluxes by a unit of aerosol optical depth. We would like to further clarify that the applied linear regressions, in this context (in section 3.4), do not aim to provide an estimation of aerosol DREs from AOD values. Instead, the objective was to determine, through the computed slopes of linear fits, the nature and extent of the dependence of DREs on AOD over the Mediterranean basin. This has been clarified in the revised text, at the end of the first paragraph of section 3.3, referred to as sect. 3.4 in the ACPD.

In conclusions, I did not find in this manuscript any piece of information that would improve our knowledge about the aerosol radiative forcing in the Mediterranean basin.

The reasons for which we believe that the present study is a significant contribution to the existing knowledge on the topic have been fully explained above. Moreover, at the beginning of his review, page C13846, lines 5-8, the Referee mentions that "*The work by Papadimas et al. is original because their radiative transfer model uses input data derived from measurements rather than aerosol fields obtained from models. This is indeed a valuable effort to approach true values.*"

In case the authors would consider re-submitting a thoroughly revised version of this manuscript, I have listed below a few specific comments, omitting missing words and spelling mistakes though.

An effort was made to correct the manuscript for missing words and spelling mistakes. Also, answers are given below to the Reviewer's specific comments.

Specific comments

- References for MODIS and GADS input data are missing.

References were provided in the Introduction (page 3, lines 102 and 104) as well as in section 2.2 (page 5, lines 177-178).

- At which altitude was the TOA set?

TOA is set at 50mb, essentially corresponding to the top of troposphere. This is because our study focuses on radiative effects of tropospheric aerosols. This has been clarified in the text (Introduction, page 3, lines 109-110).

- Parag. 3.2.1. This is an example where redundancies make the overall demonstration obscure. Sentences 2 and 3 are confusing and should be deleted. Discussing figures is more efficient than general statements.

Actually, by those two sentences, we are just starting to discuss the results of Figure 1. We believe that these sentences are necessary to: (i) introduce the reader, especially the one who is not familiar with the topic, to the meaning/nature of DRE results, and (ii) avoid potential confusion related to the positive and negative signs of DRE values, according to their determination formulas, which vary from one study to another in the literature.

- Parag. 3.2.1. As an example, this section should have concluded if the regional variations in aerosol forcing at the top of the atmosphere are due to differences in aerosol loadings, optical properties, or surface albedo (not talking about peculiar areas like the Alps or the Sahara).

Answered (see reply to main comment). Moreover, we believe that, apart from "normal" conditions of aerosol DREs, it is worth to discuss patterns associated with aerosol induced planetary warming over the specific world areas, which are relevant to climate change.

- *Page 3020, lines 6-11: units are swapped.* Fixed (page 9, line 314).

- Parag. 3.2.2. First sentence is meaningless, because it is not specific enough. Should be deleted.

We believe that the specific sentence is not meaningless, since it provides an overall qualitative and quantitative assessment of the spatial distribution of DRE_{atm} in the studied region, and thus we prefer to keep it.

- Page 30021, line 11: and partly to what?

The other reasons for the large DRE_{atm} values over the Middle-East and North Africa are high amounts of solar radiation and aerosol (dust) loadings, and relatively small cloudiness and precipitation. This was indicated in the text, page 10, lines 346-348.

- Page 30021, line 23: are large DREatm in central and northern Europe due to big aerosol optical depth or small single scattering albedo?

They are due to both. AOD values there are relatively high, up to 0.2-0.25, whereas those of single scattering albedo are relatively low, down to 0.85-0.9. The values are given now in the text (page 12, lines 417-419).

- Section 3.3: The absorbing character of aerosols is described by its single scattering albedo. How can the ratio TOA (DRETOA) / (DREnetsurf) tell more on this?

The ratio $DRE_{TOA}/DRE_{netsurf}$ is a good indicator of aerosol absorption. Of course, aerosol single scattering albedo is another indicator, but it just describes the absorbing efficiency of aerosols, and not the actual one which is better described by the specific ratio that is based on radiative quantities. This was indicated in the text, page 11, lines 386-388.

- Page 30023, lines 2-12: this is all too speculative and little convincing. The role of desert dust in low (DRETOA) / (DREnetsurf) ratios is not confirmed by the spatial variation (Fig 2i) which shows most of the <0.2 values in the northern part of the domain.

We agree with the Reviewer that attributing the low (<0.2) $DRE_{TOA}/DRE_{netsurf}$ values in the northern part of the domain to dust aerosols is not correct. However, we would like to note that we are discussing the ratio values in terms of regional averages. Of course over northern European areas, but also over the continental European areas as a whole, the low ratios are also due to absorbing aerosols, which are mostly emitted by anthropogenic activities, as already reported in section 3.2.2. The relevant text has been corrected accordingly.

- Page 30024, line 10: the "strong dependence" should be specified (slope or regression coefficient)?

Actually, it refers primarily to the slope values but also to the correlation coefficients. This has been clarified in the revised text.

- Page 30026, line 15: is the seasonality weak, or the forcing itself?

It is the forcing (DRE_{TOA}) that shows weak seasonal variation. The relevant text was rewritten to avoid confusion.

- Page 30027, line 28 (and elsewhere): "solar brightening" is not the correct concept.

The terms "solar brightening" and "solar dimming", referring to increases and decreases of surface solar radiation produced by aerosols, were removed from the text.

- *Page 30028, line 9: what about reduction in aerosol and aerosol precursor emissions?* The reductions in aerosol and aerosol precursor emissions were added in the text as potential factors explaining the decreasing regional aerosol loading and DRE_{netsurf}.

- Page 30030, lines 13-16: calculating the effect of 10% change in the aerosol optical parameters before stating that such changes "are quite difficult to take place" is another example of confusing thought process. Are these 10% changes in aerosol optical propertiesvisible in the input data or not? Why not starting straight with the computation for 1, 3, 5, 7, and 10% error then?

The estimation of changes to model aerosol DREs arising from changes in AOD, ω_{aer} , and g_{aer} by $\pm 10\%$ was chosen to quantify the relative sensitivity of DREs to these key aerosol radiative properties. This is now explained in the text to avoid confusion.

- Page 30031, line 28 and following sentences on next page is too vague for a conclusion.

We do not really understand to what exactly the Reviewer's comment refers to. If it refers to: "... According to our results, the seasonal variation of aerosol direct radiative effects is driven by the available solar radiation fluxes under both clear- and all-sky conditions. The role of aerosol loading, i.e. AOD, is important mostly in cloudless skies.", i.e. to the end of 2nd paragraph of section 5, then we believe that this conclusion arises from our findings showing that: "The maximum absolute DRE values are computed for summer (July) and the minimum ones for winter (December). However, under clear-sky conditions secondary DRE maxima appear in spring (April) in line with a corresponding spring AOD maximum."

- Page 30032, line 6: polluted aerosol is a weird concept.

The text was modified to: "...relatively high values are calculated over central and northern Europe, associated with anthropogenic emissions under conditions of atmospheric pollution."

- Page 30033, line 4: it is stated earlier (p. 30028, line 5) that cloudiness and precipitation in the Mediterranean basin increased over 2000-2007 yet.

Earlier, i.e. in section 3.4, page 16, lines 548-550, it was reported that according to satellite data, cloudiness has increased over the Mediterranean from 2000 to 2007, along with total precipitable water. No reference was made to Mediterranean precipitation changes. On the other hand, in Conclusions, page 20, line 692, through page 21, line 698, a reference is made to potential future cloud and precipitation changes, based on the aerosol induced surface and atmospheric radiative changes that were determined in the present study. Certainly, the formation of clouds and precipitation, and the assessment of future desertification over the Mediterranean basin, also including evaporation, are very complicated processes, which are driven not only by aerosols, but also by other parameters. Such processes also have responses that require some time to act with visible results, if any, while can undergo counterbalancing effects from various parameters. Therefore, admittedly, making such speculations on the role of future climate changes can be problematic, and therefore the text has been re-written.

- *Page 30033, line 5: the sentence of computed data validation is out of place.* The sentence has been moved to the first paragraph of section 5.