Reply to Prof. Dulac's Comments

We appreciate the reviewer's thoughtful comments and suggestions. They certainly improved the scientific quality of the manuscript. We have revised the manuscript as per the reviewer's suggestion. The replies of all the comments raised by reviewer are given as follows:

This paper combines remote sensing data on aerosol properties and radiative calculations in the eastern Mediterranean basin to in order to classify aerosol types encountered and ultimately derive respective atmospheric heating rates. Authors have followed my methodological suggestions made on an earlier version of their ms. for using abundant AERONET data from the Mediterranean region rather than from distant regions. I find the paper sound, clear, and appropriate for publication in ACP. I recommend publication with a minor revision, and I also suggest attachment of this paper to the recently opened ChArMEx special issue focused on chemistry and aerosols in the Mediterranean. My detailed comments are listed below.

Response: We are thankful to the reviewer's comments on our earlier version. We are happy to link this paper to ChArMEx special issue. Responses of the detailed comments are given point-by-point below.

In reply to anonymous referee #1, I need to say that in my initial evaluation I have recommended authors who were addressing all season to rather focus on the summer season, because there are much less AERONET level-2 absorption data during other seasons (see Fig. 5 in Mallet et al., ACP, 2013). In any case, this point is worth to be mentioned. I guess that only Spring might possibly offer reasonable enough statistics for further seasonal computations.

Response: Thanks for suggestion. We have mentioned this point in the revised manuscript. We have also pointed out this fact in response to other reviewer's comments.

Main issue:

My main critical comment results from the fact that radiative computations are made in the 0.25-20 um domain when AERONET observations used cover only the visible and near-infrared wavelength range (roughly 0.4-1 um): it should be clarified how aerosol properties are defined outside of the AERONET spectral range. This lack of observations in the infrared probably adds significant uncertainties, especially for large dust particles that both significantly scatter and absorb in the infrared. In another coming paper of the ChArMEx special issue, Sicard et al. (Estimation of mineral dust longwave radiative forcing: sensitivity study to particle properties and application to real cases over Barcelona, ACPD, 2014) compare the few existing papers describing the spectral dependence of the complex refractive index of mineral dust in the infrared (Volz, 1973 and 1983, Hess et al., 1998) and show that there are significant differences in the IR atmospheric window.

Response: We appreciate the critical comment raised by reviewer. In the earlier version, we extrapolated our SSA, ASYM and AOD for the entire wavelength using log extrapolation for AOD and liner extrapolation for SSA and ASYM, as was also done in previous studies. However, follwoing Sicard et al. (2014, ACPD), it seems that our LW results underestimate the forcing, as our interpolation (extrapolation) scheme is not up to mark. Therefore, we used the AERONET derived particle size distributions and refractive indices (0.4-1.0 µm) to estimate the aerosol optical properties in entire wavelength region (0.25–20 µm). The detailed methodology, which is included in revised version, is as follows:

"To perform aerosol radiative forcing calculations in 0.25 – 20 µm, aerosol properties in entire wavelength region (0.25 – 20 µm) are necessary. Since the measured AERONET aerosol optical properties are only available in the visible and near-infrared wavelength range (~0.4 – 1.0 µm), we used AERONET observed particle size distributions and refractive indices (0.4-1.0 µm) to estimate the aerosol optical properties in the entire wavelength region (0.25 – 20 µm). To extrapolate the refractive indices, we assume that the three aerosol types (dust, polluted dust and polluted continental) are internal mixtures of components with known short-wave and long-wave refractive indices. As mixing rule relating the refractive indices of mixture and components, we used the volume averaged refractive index mixing rule. The components assumed are: mineral dust and water for dust dominated aerosol; mineral dust, black carbon and water for polluted dust; ammonium sulphate, black carbon and water for polluted continental aerosol. In the latter case, ammonium sulphate is representative for various components with similar refractive indices. The refractive indices of the components are taken from Hess et al. (1998) for black carbon and mineral dust (SW), Rothman et al. (2005) for ammonium sulphate and water and I. N. Sokolik (unpublished data, 2005) for mineral dust (LW). The volume fractions are chosen such that the refractive indices integrated over the wavelengths range of the observations (440 nm - 1020 nm) agree with the observed AERONET values. We obtain the following mean volume fractions: 79.6 % mineral dust, 20.4 % water (dust); 38.5 % ammonium sulphate, 1.7 % black carbon, 59.8 % water (polluted continental); 60 % mineral dust, 0.5 % black carbon, 39.5 % water (polluted dust). Using these volume fractions combined with the refractive indices of the components and the observed particle size distributions, we compute the aerosol optical properties. SCATTNLAY (Peña and Pal, 2009) Mie code is employed for calculations of optical properties (AOD, AAOD, SSA, ASYM). To obtain an error estimate, the standard deviation of the observations is propagated using jackknife resampling (Wu, 1986). The output AODs for each aerosol types is scaled with CALIOP-derived AOD."

New results of SSA, ASYM and AOD for entire wavelength (0.25- 20 µm) are given in Fig. R1. We found a significant increase in LW forcing for dust and polluted dust aerosols after these corrections. Fig. R2 and Table R1 strengthen the point raised by reviewer. We are thankful for this comment. We included all these figures and detailed methodological corrections in revised manuscript. We revised the manuscript (in terms of forcing calculation) as per the new results.

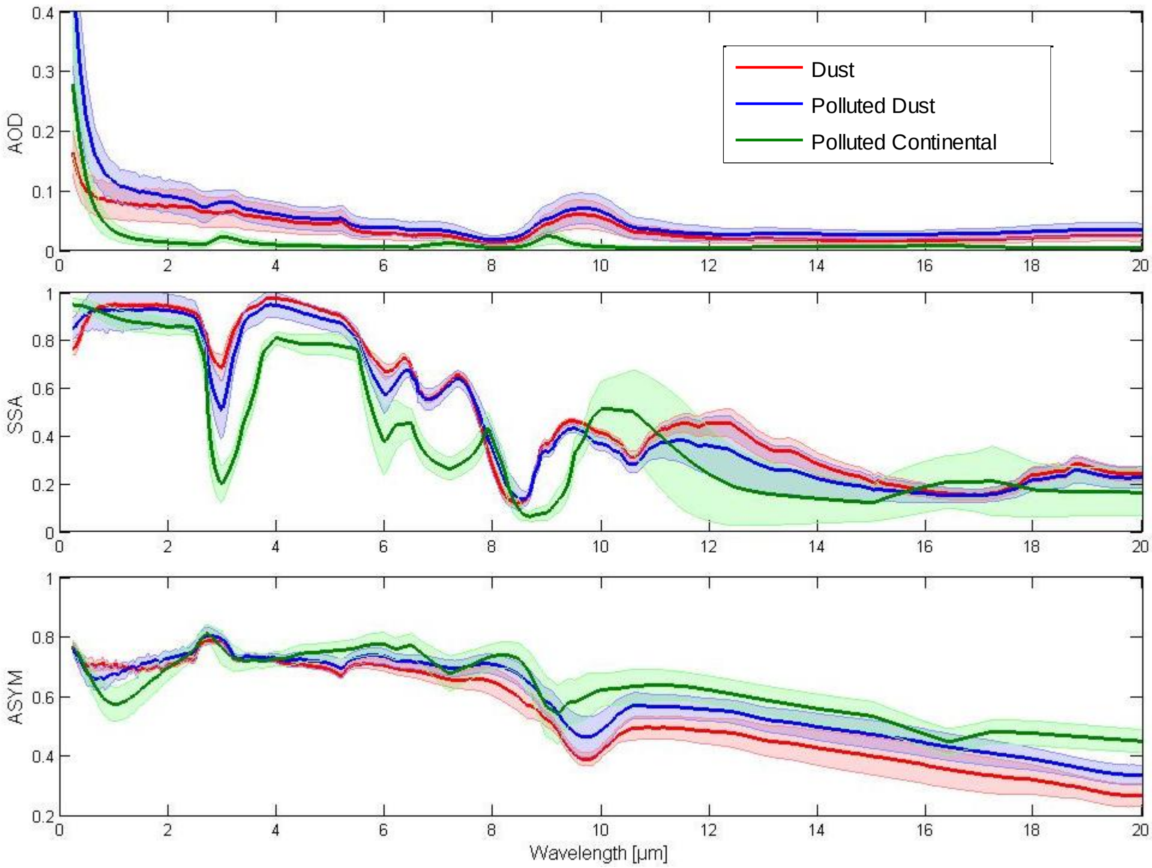

Fig. R1 AOD, SSA and ASYM for three different aerosol types in 0.25 – 20 µm wavelength region. The errors in calculation are shown by transparent shaded area.

Table R1 Day-time average aerosol radiative forcing [Wm^{-2}] in short wavelength (SW) and long wavelength (LW) region for different absorbing aerosols during summer 2010 over the ROI in the Eastern Mediterranean basin.

Aerosol Type	**SW (0.25 - 4.0 µm)**		**LW (4 - 20.0 µm)**	
	$(\Delta F_{aer})_{SRF}$	$(\Delta F_{aer})_{TOA}$	$(\Delta F_{aer})_{SRF}$	$(\Delta F_{aer})_{TOA}$
Dust	-21.95	-11.05	2.47	0.91
Polluted Dust	-39.95	-20.60	3.31	0.64
Polluted Continental	-17.41	-10.27	0.78	0.07

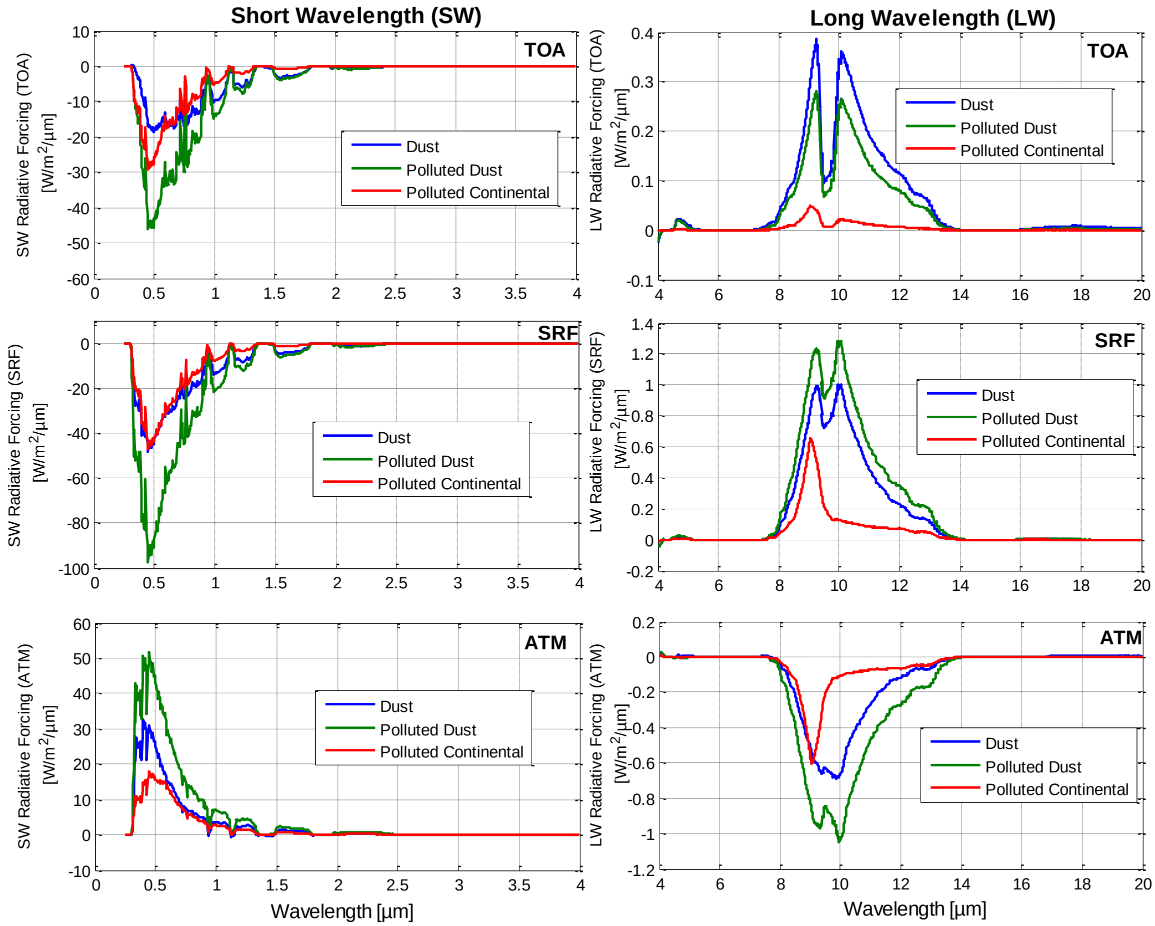

Fig. R2 Radiative forcing of different aerosol types as a function of wavelength in SW and LW region at TOA, SRF and in ATM.

Minor comments:
-Surface albedo is an important parameter in radiative forcing computations, which seems not addressed here: some details should be provided.

Response: Thank you for mentioning this point. We included some details about surface albedo used in our calculations. **"SBDART characterized 'ocean water' surface type was used to parameterize the spectral albedo of surface (Tanré et al. 1990; Ricchiazzi et al., 1998)."**

-Clarification of the aerosol classification methodology would be welcome (top of p.2409). I suggest:"[...] over the Mediterranean. We have classified aerosol events based on individual (or daily?) AERONET observations. Our classification [...]. For aerosol classification as dust, we used [...]. For classification as polluted dust [...] we included [...] other dust dominated sites and selected AERONET data with $0.7<EAE<1.1$. Pollution[...]".

Response: We corrected accordingly.

-I recommend an additional figure S1b showing the polluted continental aerosol case (0.7<EAE<1.1).

Response: We added Fig. S1b for polluted continental aerosol cases.

-Section 3.1, p.2410: I do not think that it can be reasonably argued that the difference in AOD between MODIS and MISR is due to a difference between their respective wavelengths of 550 and 555 nm (item 3): even with an EAE as high as 2.2, the difference in AOD would only be of 2%.

Response: We deleted this sentence from the revised manuscript.

-Section 3.1, p.2411: the (by far) highest value of AAE at Lampedusa Island (2.24) is questioning since Blida in North Africa shows a lower value of 2.02. To my knowledge, a value of 2.24 is unusually high, even for sites in dust region: explanation deserves to be left more open.

Response: We added one sentence after the explanation, which makes it open for more introspection. **"However, the explanation of this high AAE value of Lampedusa needs more deep introspection."**

-Section 3.1, p.2412, 1st paragraph: you might comment the summer means (Table 2) compared to overall means (Table S1) and the role of dust in summer.

Response: We added some insight about comparison of both tables (EAE values) and have commented about the role of dust in summer season. The added part is as follows: **"The role of dust in summer could be seen by comparing the summer means (Table 2) and overall means (Table S1) of EAE for dust affected sites situated in western basin (Blida, Malaga, Granada etc.). Dust events during summer are likely to decrease the EAE values over these sites. However, the eastern basin sites are more influenced by pollution as seen from increased values of EAE in summer as compared to overall means."**

-Section 3.1, top of p.2416: you might also consider Ramanathan et al. (JGR, 106, 28371-28398; see plate 18), who report computations of seasonally averaged heating rates over the Indian Ocean due to anthropogenic carbonaceous aerosols, and Zhu et al. (JGR, 112, doi:10.1029/200JD008427; see Fig. 11), who report heating rates of mineral dust over various marine regions.

Response: Thank you for providing these interesting references. We added the results of these papers in our revised manuscript. **"Ramanathan et al. (2001) has reported seasonal (JFM, 1999) and vertical averaged (0-3 km altitude) heating rate of ~0.3-0.6 K/day due to anthropogenic carbonaceous aerosols over the north Indian Ocean region. Mineral dust layers have also shown heating rate of about 0.5 K/day over the Arabian Sea and the Sahara coasts (Zhu et al., 2007)."**

-Section 3.3: you might wish to discuss the fact that the temperature lag between 925 and 850 hPa is constant whatever the AOD.

Response: Thank you for this important observation from Fig. 10. We have discussed this point in our revised manuscript. We have also provided a supplementary figure for the explanation of this trend. **"Fig. 10a shows that the difference between the temperatures at 850 hPa and 925 hPa is independent of aerosol loading, i.e. it is almost constant in the entire range of AOD (0.07 to 0.58). This observation suggests that the contribution of absorption from these two aerosol layers (at 850 hPa and 925 hPa) is almost similar in magnitude. Fig. S5 strengthened our abovementioned conclusion that the maximum observation due to absorbing aerosols (dust, polluted dust and polluted continental) occurs between ~400 and ~2200 m altitude range with almost similar relative frequency of occurrence."**

-Some figures are difficult to read, see relevant technical comments hereafter.
Response: We improved all our figures, which were difficult to read.

Technical points:
-General: check the occurrences of a double f within words throughout the text (affect, effect, difference...): they have all been put in italic style, likely due to the use of the symbol ff for the aerosol fine fraction.
Response: Done

-Introduction, p.2405, line 8: remove "by".
Response: Done

-Introduction, p.2406, line 22: "there has been no direct measurement" (singular)
Response: Corrected

-Methodology, p.2407, line 10 and p.2408, line 22: "Derimian" with two i.
Response: Corrected

-Methodology, p.2407, lines 20-21: reorder references by chronological order.
Response: Done

-Methodology, p.2408, line 3: replace "board on" by "on board".
Response: Done

-Methodology, p.2410, line 3: provide reference for the model atmosphere used.
Response: Done

-Methodology, p.2410, line 24-25: specify that the spring peak is in April and that the winter minimum is more exactly from November to January.
Response: Done

-Results and discussion, p.2411: specify “also manifested by larger SSA440 values”.
Response: Done

-Results and discussion, p.2412, line 3: should be “Mallet et al. (2013) consider the”.
Response: Done

-Results and discussion, p.2412, line 1: specifiy “0.2-0.5) with a maximum in the SW part of the basin, whereas”.
Response: Done

-Results and discussion, p.2412, line 7: stop sentence after “variability”.
Response: Rephrased

-Results and discussion, p.2413, line 18: add “over the period 1983- 1994” at the end of 1st paragraph.
Response: Done

-Results and discussion, p.2415, line 4: correct “the probability [...] is found to reach”.
Response: Corrected

-Results and discussion, p.2415, line 11: change “from the lowest bin (0.07) to the highest (0.58)”.
Response: Done

-Results and discussion, p.2415, lines 19-20: change to “(AOD _0.08) between 1000 and 850 hPa is significantly [...] for 1400 m). This indicates the stable”.
Response: Done

-Results and discussion, p.2416, lines 7-8: change “to be comparable to” by “to interact with”.
Response: Changed

-Results and discussion, p.2416, line 15: “integrate the effect of” might be better than “represent the average measure of”.
Response: Done

-Results and discussion, p.2416, line 20: remove “;” within the parentheses.
Response: Done

- Results and discussion, p.2418, line 23: “region” rather than “regime”.
Response: Done

-Results and discussion, p.2418, line 24: “produces” rather than “produced”.
Response: Done

-Implications, p.2420, line 1: you might refer to Ackerman et al. (Science, 2000) who describe this effect for absorbing soot particles.
Response: We have included this important citation in revised one.

-Implications, p.2420, line 8: add article in "of a pollution pool". -Conclusion, p.2420, line 18: add article in "with a radiative transfer model".
Response: Corrected
-Conclusion, p.2421, line 8: specify "In summer 2010, the daytime average forcing is found [...]".
Response: Done

-References: Marconi et al. (2013) cited p.2408, Omar et al. (2009) cited in p.2409, and Xiao et al. (2009) cited in p.2410 are missing in the list.
Response: Done

-Table 1: in the legend, specify "MODIS and MISR summer-time mean AOD at 550-555 nm (±standard deviation) within".
Response: We want to clarify that Table 1 shows only MODIS derived AOD. We have rephrased the title as **"MODIS summer-time mean AOD at 550 nm (±standard deviation) within the aerosol layer over the ROI for 10 years (2003-2012)."**

-Table 3: in the legend, specify "over the ROI in the eastern Mediterranean Basin"; add a column with average AOD.
Response: We think that reviewer want to point out Table 4 instead of Table 3. We have revised Table 4 as per suggestion.

-Fig. 5 is hardly readable; please use bold lines and may be a dotted line for the green or blue line which colours are close, and enlarge to maximum size in the page.
Response: We have revised the figure as per suggestions.

-Fig. 7: filling rectangles would be helpful to the reader.
Response: Done

-Fig. 10: in the legend, specify "into equally spaced bins of 0.05 AOD550; enlarge to maximum size for the page; rescale the right axis of the bottom right plot to fit the left axis so that you can remove green symbols for plotting the numbers of occurrences.
Response: Done

-Fig. 10b and fig. 11b: use the plural for "occurrences" in the legend of the right axes.
Response: Done

** *Note: References are listed in revised manuscript.*