Reply to Reviewer #1

We are very thankful for the reviewer's comments and suggestions. It will certainly increase the robustness of the present 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:

In this study, the authors characterise the optical properties of aerosols over the Eastern Mediterranean using ground-based and satellite remote-sensing retrievals. Those optical properties are then used in radiative transfer calculations to estimate the direct radiative effect and changes to heating rates exerted by the aerosols. The results are interesting (Figure 10 particularly) and the region is well chosen for his mixture of aerosols with varying absorption properties. The scope is disappointing, however, and the authors could have easily extended their analysis over the full period, rather than just June–August 2010. Contrasting the results with the same analysis, but applied to Western Mediterranean aerosols, would also have been most welcome.

Response: We thank the reviewer for his valuable comments and suggestions. We have used only the summer season for our detailed analyses since (1) the summer season is almost cloud free over the Eastern Mediterranean and we do not want to confront with clouds radiative properties while dealing with absorbing aerosols (mentioned in manuscript), (2) another reason for choosing only the summer season is that there are much less AERONET level-2 absorption data during other seasons (Mallet et al., ACP, 2013), as also suggested by second reviewer in our earlier version.

We appreciate the reviewer's suggestion that we could extend this study to the western Mediterranean Basin as well. However, we feel that this is beyond the scope of the current study at the moment. Concerning the suggestions, we would like to see this type of comparison in our upcoming study over the Mediterranean Basin.

Nevertheless, my opinion is that the study is worth publishing, provided that the comments below are addressed satisfactorily. In particular, the choice of a single solar zenith angle, and the use of AERONET-retrieved single-scattering albedo, can both lead to an overestimate of absorption and heating rates.

Response: Thank you for the helpful comments and suggestions. We have tried our best to give the response of your major concern regarding overestimation of absorption and heating rates using a single solar zenith angle, and AERONET-retrieved single-scattering albedo.

1. Main comments

Reading the paper, one has the feeling that MODIS could have been put to more use. There are almost two independent analyses in the paper: one with MODIS (and MISR on occasion), which gives an idea of seasonality and inter-annual variability before providing an interesting correlation with AIRS-retrieved temperature profiles. The second

with AERONET and CALIOP, which provides the inputs to the radiative transfer calculations. Those two strands could be better coupled. Why not combine MODIS AODs and CALIOP vertical profiles, to give a really three-dimensional view of the situation? Why not use the estimated heating rates to analyse the results from Figure 10 more deeply?

Response: We would always like to maximize the use of MODIS data. As the reviewer rightly pointed out that, there are two different strands are used to calculate heating rate. We have tried our best to couple these two strands in the present study. Now, we have added some more observations from Fig. 10a and explained it with the help of CALIOP measurement. The added analyses are as follows:

"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 absorption due to absorbing aerosols (dust, polluted dust and polluted continental) occurs between ~400 and 2200 m with almost similar relative frequency of occurrence."

However, 'combining MODIS AODs and CALIOP vertical profiles, to give a really three-dimensional view of the situation' is difficult at this stage as CALIOP swath is very narrow as compared to that of MODIS. In our view, providing three dimensional aerosol distribution by combining MODIS AODs and CALIOP vertical extinction profiles requires a stand-alone study.

Apart from that we have also tried to compare estimated heating rates from Fig. 10. The added analysis is as follows:

"The overestimated heating rate from AIRS vs MODIS observation as compared to model calculations is attributed to the treatment of different aerosol types by the RTM, whereas the remote sensing calculation combined the effect of all aerosol types present. In addition, the different AOD used in both methods (CALIOP-derived in RTM and MODIS-derived in other one) could be an important reason for these differences."

Section 2, page 2410, line 3: Using a single, daytime-average value of the solar zenith angle neglects the strong dependence of aerosol direct radiative effects (DREs) on solar zenith angle (see Figure 5 of Boucher et al., 1998). Nowadays, a proper integration of instantaneous DRE calculated at regular intervals throughout the day is easy to do, and prevents DRE from being overestimated by the choice of a zenith angle that is close to the maximum instantaneous DRE. This also allows a proper account for seasonality of DRE, which is not only driven by total insolation (as implied by the authors on Page 2411, line 6), but also by different distributions of solar zenith angle.

Response: We appreciate the reviewer for this important point regarding the choice of solar zenith angle (sza). We have re-calculated the DREs for $0^0 < sza < 90^0$ with 5^0 regular intervals. The variation of DREs with sza is certainly strong at the top of the atmosphere (TOA) and at the surface (SRF) as also mentioned by Boucher et al. (1998). Fig. R1 shows these variations for each absorbing aerosol types included in this study. We have also compared the cosine of sza weighted mean DREs with that from $sza=60^0$ (Fig. R2). The reviewer was correct that using $sza=60^0$ would lead to overestimation of the forcing at TOA and at the SRF. Our results suggest that choosing single $sza=60^0$ overestimates the forcing at SRF and at TOA, but it does not overestimate the aerosol forcing in atmosphere (ATM). In fact leads to a small underestimation (as shown in Fig. R2). We have also checked the heating rate profiles, which are almost same for $sza=60^0$ and cosine weighted mean. Therefore, we have used $sza=60^0$ to show our results for daytime averaged (an approximation) because we want to compare our model assessment with that of AIRS vs MODIS analyses, which use instantaneous observations at 1.30 PM (LT) for each day. For the clarity of manuscript, we have added the following discussion to the first paragraph of section 3.4 and added the Figures to the supplementary materials:

"The strong dependence of aerosol direct radiative forcing (ADRF) at top of the atmosphere (TOA) and at the surface (SRF) on solar zenith angle (sza) has well observed by Boucher et al. (1998). In this study, the variation of radiative forcing of aerosol types (dust, polluted dust and polluted continental) with solar zenith angle (SZA) at TOA, at SRF and in the atmosphere (ATM) are given in Fig. S7 (supplementary material). Our results agree with those of Boucher et al. (1998). In spite of the strong variability of ADRF (at TOA and SRF) with sza, we used $sza=60^0$ to show our results for daytime average (an approximation) because we compare our model assessment with the AIRS vs MODIS analyses, which use instantaneous observations at 1.30 PM (LT) for each day. It is worth mentioning that using a single $sza= 60^0$ (that is close to the maximum instantaneous DRE) overestimates the calculated DRE as compared to cosine of sza weighted mean DRE at TOA and at SRF, but in the atmosphere (ATM) it will be slightly underestimated (Fig. S8)."
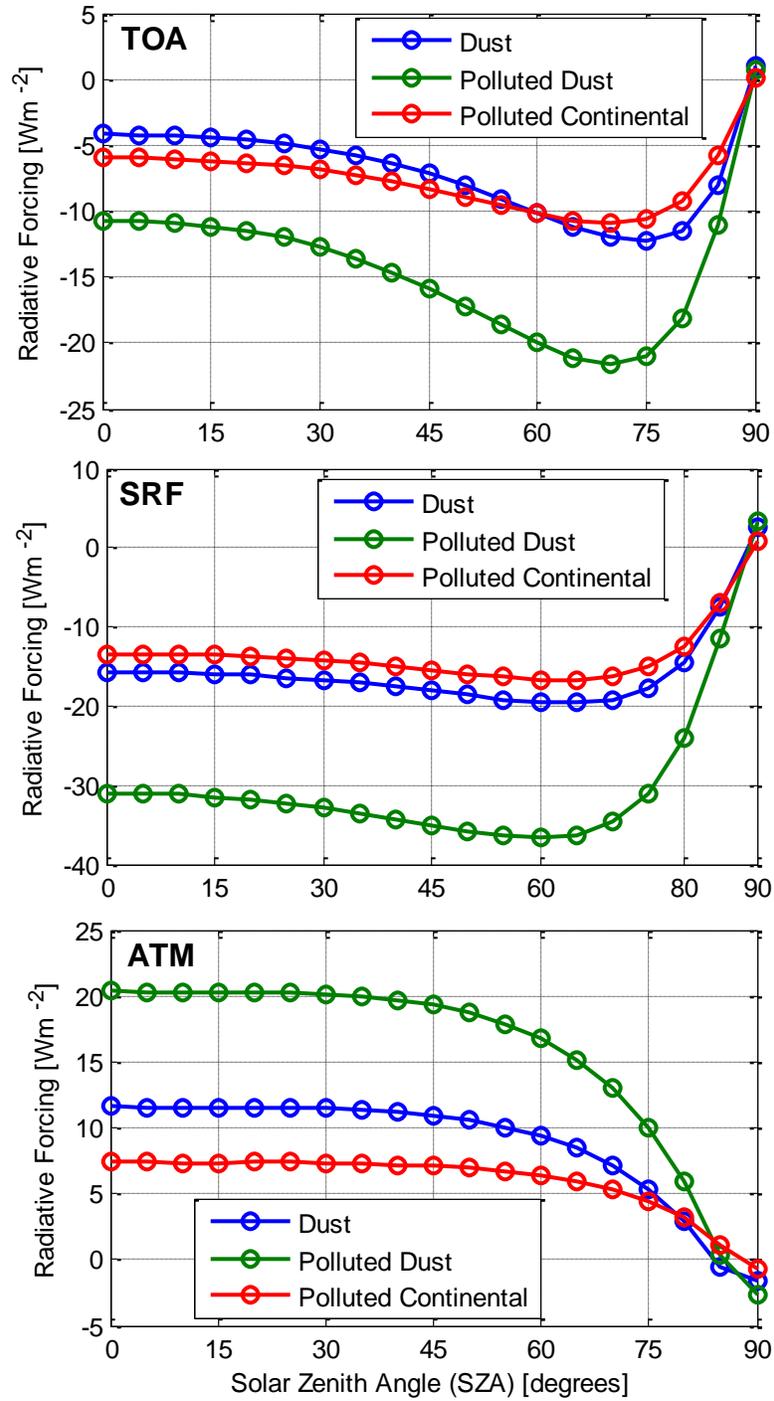
Fig. R1 Variation of radiative forcing of aerosol types (dust, polluted dust and polluted continental) with solar zenith angles (SZA) at TOA (upper panel), at SRF (middle panel) and in ATM (lower panel).
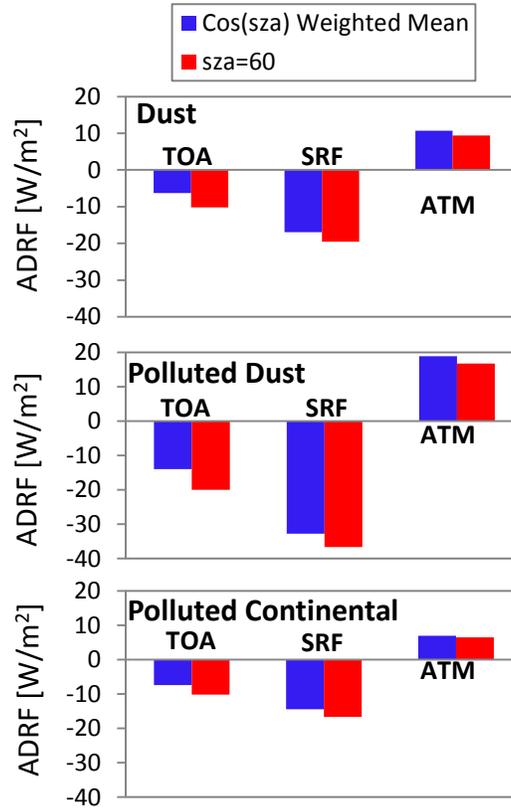
Fig. R2 Comparison of Aerosol Direct Radiative Forcing (ADRE) for sza=60^0 with Cos(sza) weighted mean for all three absorbing aerosols types at TOA, at SRF and in ATM.

Table 2: The AERONET single-scattering albedo and size distribution are only retrieved in specific conditions. One particular requirement is that the AOD is large enough (typically larger than 0.2, see section 2a of Dubovik et al., 2002). Level 2 data should take care of that problem (although it is worth checking), but that does mean that retrievals are only representative of thicker aerosol plumes. If those plumes are associated with more absorbing aerosols (mineral dust or large pollution events), then the authors use a single-scattering albedo that is biased low, and overestimate the atmospheric absorption and heating rates.

Response: Reviewer has rightly pointed out that AERONET-level 2 SSA data is only retrieved in case of higher AODs. In this context, we would like to make a point that, "SSA is an intensive property that is solely determined by the chemical composition and size distribution of the aerosol mixtures, rather than extensive property that is governed by the amount of aerosol mass loading". We have characterized our aerosol types on the basis of size parameter and source regions and we have sampled only those SSA values. Similar type of approach has been by Bahadur et al. (2011), where they have calculated AAOD for low AOD cases using the same SSA values. Therefore, SSA retrieved from thicker aerosol plumes (in case of AERONET observation) could be well representative

of thin aerosols plumes of same chemical composition and size distribution. In this study we have used AERONET-derived aerosol properties as a reference for each aerosol classes. As per the suggestions by the other reviewer, we recalculated the aerosol optical properties for the entire wavelength range (0.25-20 μm) using AERONET-retrived particle size distributions and refractive indices (0.4-1.0 μm) for each class. The re-written methodology is as follows:

"To perform aerosol radiative forcing calculations in the 0.25 – 20 μm wavelength range, aerosol properties in the 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."

Section 3.3, page 2414, lines 10–18 and Figure 9: The discussion of Figure 9 is unclear. What exactly constitutes the "close agreement" claimed by the authors? Are MODIS fine-mode fractions compared to the full CALIOP cross-section, or only the parts highlighted by the black circles? This is important because, if I understand the paper correctly (but clarify the text if I'm wrong), CALIOP's classification is used directly to obtain the profiles shown on Figure 8b.

Response: The Reviewer is correct that CALIOP's classification is used directly to obtain the profiles shown on Figure 8b. We are sorry for unclear explanation of Fig. 9. We compared the MODIS fine fractions (*ff*) to only the parts highlighted by black circles

(ROI region) [now this is clearly mentioned in the figure caption]. Cornering the fact that in our each case marine aerosols are present which will likely decrease average *ff* values, average values of *ff* found in each case are in close agreement with the 'mentioned studies'. Now we have paraphrased the entire paragraph as follows:

"**Fig. 9 shows the assessment of MODIS *ff* for three different scenarios of dominant aerosol loading (dust, polluted dust and polluted continental) over the ROI. The mean values of *ff* are 0.44±0.11, 0.55±0.12 and 0.64±0.17 for the dust, polluted dust and polluted continental dominant aerosol classes, respectively. Earlier studies have reported MODIS *ff* in the range of 0.25 – 0.45 for marine aerosols, 0.37 – 0.51 for dust, and 0.83 – 0.92 for anthropogenic aerosols over the various oceanic regions of the world (Kaufman et al., 2005; Yu et al., 2009; Jones and Christopher, 2007; 2011). Relatively lower values of *ff* are found in our cases (as compared to above-mentioned studies). These could be understood as marine aerosols are present in all three cases in our study (Fig. 9, black circles), which would likely decrease the average *ff* values for all three cases. Therefore, distribution of *ff* as compared to other above-mentioned studies is in close agreement with the aerosol classification for this present study.**"

2. Other comments

Abstract, page 2404, line 4: "model calculations" –> "radiative transfer model calculations"
Response: Corrected

Figure 3: It would be useful to take the standard deviations (denoting variability) from Table S1 and show them as whiskers on the Figure. That would provide an efficient graphical summary of the AERONET dataset.
Response: Thanks for suggestion. We have shown the standard deviation by whiskers on the figure.

Introduction, page 2405, line 16: "temperature increase": surface temperature?
Response: Sorry for confusion. We have corrected the sentence. **"…..can lead to a temperature increase of that layer by 2-4 K"**

Section 2, page 2410, line 9: Clearly state that radiative transfer calculations cover both the shortwave and longwave spectra. For longwave calculations, how is the temperature of the aerosol layer given? Is it that of the corresponding standard atmosphere level, or does it include any absorption-driven warming of the aerosol layer? Also, the text should note that radiative transfer calculations are done in cloud-free sky.
Response: We have clarified the point raised by reviewer that we have performed the solar irradiance calculation covers both SW and LW in clear sky conditions. We appreciate the reviewer's comments regarding the use of temperature profile in LW region. We understand that it will be more appropriate if we include absorption-driven warming of the aerosol layer to the temperature profiles. However, we have used the

corresponding standard atmosphere level in LW region, which could be understood as our limitations.

Figure 4: Clearly state what the standard deviation measures: interannual variability? Spatial variability? Retrieval uncertainty?
 Response: Done, "standard deviation measures inter-annual variability"

Section 3.3, page 2415, line 12: "no statistically significant changes at 1000 hPa". Does that mean that temperature changes are statistically significant at the lower pressure levels? At which confidence level?
Response: We have used "no statistically significant changes" in terms of standard deviation. Now we have clarified it in text.

Section 3.3, page 2416, lines 7–8: "Dust particles are large enough (up to several micrometer) to be comparable to IR wavelength" is an awkward thing to say. I would suggest "Because of their large sizes (up to several micrometers), dust particles are able to strongly interact with IR radiation."
Response: Corrected

Section 3.3, page 2416, lines 19–20: "plausible reason have been explained in the Supplement". The explanation is short enough to be reproduced in the main text, which would remove the need to consult the caption of Figure S4.
Response: We have included this reason in the last paragraph of section 3.3 in the revised version .

Section 3.3, page 2417, line 12: The relative strength of uncertainties merits a discussion. Are some uncertainties weaker because of compensating errors between AOD and SSA?
Response: We are sorry for putting these uncertainties values in terms of %. The large differences in uncertainties are found because we have calculated in terms of %. For example, 6.8 Wm^{-2} error in case of polluted dust forcing (-36.7 Wm^{-2}) at SRF will produce about 18% uncertainties, whereas 6.0 Wm^{-2} error in case of polluted continental forcing (-16.7 Wm^{-2}) at SRF will produce about 36% uncertainties. In order to avoid confusion, we have deleted that part from the paper.

Conclusion, page 2421, lines 10–14: This paragraph is over-enthusiastic. The "detailed assessment" is in fact the speculative, two-paragraph section 4. The wording should be toned down here. The authors should wait for the results of their EMAC study before making such claims.
Response: We re-phrased the sentence.

3. Technical comments

Introduction, page 2405, line 8: "budget by both directly" –> budget both directly
Response: corrected

Section 2, page 2408, line 3: "is board on" –> is on-board

Response: corrected

Section 2, page 2410, line 1: Typo: McComiskey
Response: Corrected

Please improve the quality of Figures 5, S1, S3, S4, and S5 by plotting thicker lines.
Response: All the figures have been revised

Typo in Figure S2: "Uncertainty"
Response: corrected

Section 3, page 2410, line 25: "means" –> mean
Response: corrected

Section 3, page 2411, line 17: "the North Africa" –> North Africa
Response: corrected

Section 3, page 2411, line 19: Sede Boker
Response: corrected

Section 3, page 2412, line 10: "and mixes" –> that mix
Response: corrected

Section 3.2, page 2413, line 7: "increase" –> increases
Response: corrected

Section 3.4, page 2417, line 11: "rage" –> range
Response: corrected

References:
Bahadur, R., Praveen, P. S., Xu, Y., & Ramanathan, V. (2012). Solar absorption by elemental and brown carbon determined from spectral observations. *Proceedings of the National Academy of Sciences*, *109*(43), 17366-17371.

**Note: Other references are listed in revised manuscript.