

***Interactive comment on* “The direct effect of aerosols on solar radiation over the broader Mediterranean basin” by C. D. Papadimas et al.**

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We would like to thank the Reviewer for the useful comments that helped us to improve our manuscript. We have taken into account all suggestions and addressed the raised issues trying to provide necessary clarifications and improvements. Below are given point by point answers to the comments (also provided in *Italics*).

General Comments

- as often found in the literature, radiative effects are calculated from monthly average aerosol optical properties. However, aerosol optical properties change at a much faster rate, and an accurate calculation should take into account its high temporal resolution evolution. Calculations of the monthly average radiative effect which use monthly

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Discussion Paper

average aerosol optical properties imply that the radiative effects depend linearly on changes in the aerosol optical properties (although the progressive changes of the solar zenith angle daily course and clouds should tend to produce non linearities). The analysis in section 4 of the paper suggests that for small changes of the optical properties the radiative effect responds linearly, and it is probably possible to proceed by using monthly average aerosol properties. However, the behaviour may be non linear for larger changes; and changes in the wavelength dependence of AOD (i.e., of the Angstrom exponent; see also comments below) are not taken into account. A comment on this aspect, and on possible additional associated uncertainties, should be added.

First, we would like to emphasize that our RTM takes into account the wavelength dependence of AOD, ω_{aer} and g_{aer} . As noted in the 4th paragraph of the Introduction “The RTM uses as input spectral aerosol optical properties (AOD, asymmetry parameter, g_{aer} , and single scattering albedo, ω_{aer}) taken mainly from the MODerate resolution Imaging Spectroradiometer (MODIS, King et al., 2003; Remer et al., 2005) of NASA (National Aeronautics and Space Administration) and supplemented by the Global Aerosol Data Set (GADS, Koepke et al., 1997)”.

In the present study, the direct radiative effect of aerosols over the Mediterranean basin is derived on a monthly basis, based on RTM model computations using monthly mean input aerosol optical properties, but also other surface and atmospheric parameters, e.g. clouds. The specific method can be considered as appropriate and computationally cost efficient when aerosol DREs are to be examined on a monthly mean basis to provide climatological assessment. However, we agree with the Reviewer that there are two issues that need to be critically addressed in the revised version of our manuscript. The first is the evaluation of the day-by-day variability in the DREs due to the short term changes in the aerosol optical properties. The second is the uncertainty in the calculations based on the monthly mean AOD instead of averaging the DREs calculated on a daily basis.

The day-by-day variability, mentioned by the Reviewer, is produced by the significant day by day variability of AOD, single scattering albedo (ω_{aer}) and asymmetry parameter (g_{aer}). To evaluate this variability of the aerosol DRE components, i.e. $\Delta(\text{DRE}_{\text{TOA}})$, $\Delta(\text{DRE}_{\text{atm}})$, $\Delta(\text{DRE}_{\text{surf}})$ and $\Delta(\text{DRE}_{\text{netsurf}})$, due to changes of AOD (ΔAOD), ω_{aer} ($\Delta\omega_{\text{aer}}$) and g_{aer} (Δg_{aer}) we performed additional sensitivity studies with our RTM, similar to those presented in section 4 and Fig. 7. These simulations show a linear dependence of DREs on changes in AOD even for changes of aerosol optical properties as large as 50%. We have further quantified the day-by-day variability of the three aerosol optical properties, i.e. ΔAOD , $\Delta\omega_{\text{aer}}$ and Δg_{aer} on the DREs. Based on the daily MODIS AOD data, the day-by-day variability of AOD is found equal to $\Delta\text{AOD}=0.048$. The changes in the DREs induced by this ΔAOD are calculated to be $\Delta(\text{DRE}_{\text{TOA}}) = 0.064 \text{ W m}^{-2}$, $\Delta(\text{DRE}_{\text{atm}}) = 0.054 \text{ W m}^{-2}$, and $\Delta(\text{DRE}_{\text{netsurf}}) = 0.064 \text{ W m}^{-2}$. Compared to the computed average values of DREs ($\text{DRE}_{\text{TOA}} = -2.4 \text{ W m}^{-2}$, $\text{DRE}_{\text{atm}} = 11.1 \text{ W m}^{-2}$, and $\text{DRE}_{\text{netsurf}} = -13.5 \text{ W m}^{-2}$, the corresponding variabilities are small ($<3\%$).

A similar procedure was followed in order to estimate the variability of aerosol DREs due to day by day variability of aerosol single scattering albedo and asymmetry parameter as derived from the MODIS daily data. The estimated variability of single scattering albedo is $\Delta\omega_{\text{aer}}=0.01$, introducing, according to the derived formulas (see Figs. 7i-b, 7ii-b, 7iii-b), variabilities in the DREs of $\Delta(\text{DRE}_{\text{TOA}}) = 0.6 \text{ W m}^{-2}$, $\Delta(\text{DRE}_{\text{atm}}) = 1.6 \text{ W m}^{-2}$, and $\Delta(\text{DRE}_{\text{netsurf}}) = 0.6 \text{ W m}^{-2}$ that correspond to 25%, 14.4%, and 4.4% of the computed average DREs at TOA, in the atmosphere and at the surface, respectively. Following the same procedure, the variability of aerosol asymmetry parameter is $\Delta g_{\text{aer}}=0.03$ and the estimated DRE variabilities are equal to 12.5%, 0.002%, and 0.9% at TOA, in the atmosphere and at the surface.

The uncertainties introduced in our calculations by the use of RTM monthly mean input data, in the model radiative flux calculations, and hence aerosol DREs have been extensively investigated in earlier studies (i.e. Hatzianastassiou et al., 2005; Matsoukas

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et al., 2005). These earlier works investigated the difference between the “monthly mean fluxes” calculations based on the monthly mean input, such as those used in this paper, and the monthly-mean fluxes averaged from hourly/daily fluxes using the same RTM. Extensive comparisons have shown very good agreement, with a correlation coefficient equal to 0.985. Moreover, this problem is alleviated by the fact that the monthly mean model input data, e.g. ISCCP-D2 cloud properties, are based on a radiatively linear average method, so that differences are small.

As suggested by the Reviewer, a paragraph discussing the uncertainties of model aerosol DREs (e.g. see next comment) is added in section 5 of the revised manuscript.

- it is not clear to me how the aerosol optical properties from GADS and MODIS are combined. The GADS dataset provides a broad seasonal (winter and summer) and spatial distribution of the aerosol properties, while spatially distributed monthly mean values are derived from MODIS. How the two datasets are integrated? How is the spectral single scattering albedo associated with the observed optical depth? Is there any aerosol type attribution, to associate the GADS single scattering albedo with the MODIS AOD?

The aerosol optical properties (AOD, ω_{aer} and g_{aer}) used as model input data are described in section 2.2. As explained there, spectral AOD and g_{aer} data are taken from the MODIS database whereas ω_{aer} data are taken from GADS. The treatment of GADS ω_{aer} data is detailed in the papers by Hatzianastassiou et al. (2004a and 2007a) to which reference is made in the present paper (section 2.2, page 5, lines 184–187) in order to save space. Indeed, there is a difference in the spatial and temporal resolution between GADS and MODIS aerosol optical properties, since global distributions of GADS aerosol properties are given as climatologically averaged values on a $5\text{deg}\times 5\text{deg}$ latitude-longitude resolution for the periods December through February (northern hemisphere winter) and June through August (northern hemisphere summer). To match the spatial resolution of the climatological parameters, especially that of relative humidity to which the aerosol properties are sensitive, the original GADS

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aerosol optical properties were downscaled to $2.5\text{deg}\times 2.5\text{deg}$ latitude-longitude resolution, as explained in Hatzianastassiou et al. (2006). The aerosol properties originally taken from GADS, were re-computed for actual relative humidity values for the aerosol layer, taken from NCEP/NCAR Reanalysis project, for the study period 2000–2007. In this way, the model desired monthly distribution of ω_{aer} is consistent with the originally taken GADS seasonal ω_{aer} data. This elaboration of GADS-based ω_{aer} data in the model is now further explained in section 2.2 of the revised version.

The uncertainty in the model computed aerosol DREs introduced by this treatment of GADS ω_{aer} data is evaluated to be equal to 8%. This ω_{aer} uncertainty can induce a corresponding uncertainty of model computed DREs which, as estimated through the Equations derived in section 4 (Figs 7ii-a, 7ii-b and 7iii-b), ranges from 4.6% (for DRE_{net}surf) to 9.7% for (DRE_{atm}). These are now reported in the Discussion (section 5).

- the analysis shows that the radiative effects strongly depend on the surface albedo. This is an expected result. I would suggest that data are first grouped according to classes of surface albedo (at least ocean and desert separately), and results are discussed separately. I assume that the regional averages (tables 1, 2, S2-S5) include areas with different albedoes. I would add to the tables also the results for the sea and land separately.

Indeed, aerosol DREs significantly depend on surface albedo, as also shown in this but also some of our previous studies (e.g. Hatzianastassiou et al., 2004b). We agree with the Reviewer and follow the suggestions to classify DREs according to surface types (and albedo values). For this, we have distinguished three main surface types: land, ocean and desert. Subsequently, we grouped and computed DRE_i values for each one of the three surface types. The results, which are given in a new Table (Table 2), on an annual (7-year) and regional mean basis, reveal clearly larger DRE values over desert than over land and ocean, for all DRE components, mainly due to the larger desert dust AOD values. It is also worthy to note the positive sign of the desert DRE_{TOA} value (4.1

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W m⁻²), implying a net energy gain over deserts, opposite to cooling net energy loss over land and ocean. The energy loss has, however, smaller magnitude over land than oceans (-0.4 against -5.4 W m⁻², respectively) because of the larger surface albedo in the former case. At the surface and in the atmosphere, the magnitude of aerosol DRE is larger over land than over ocean. Appropriate discussion of Table 2 has been added in the revised manuscript.

- the derived radiative effects are discussed throughout the paper in terms of AOD, surface albedo, single scattering albedo, and daytime duration. I believe that the spectral dependence of the aerosol optical depth (or the Angstrom exponent) plays an important role in modulating the radiative effects. As discussed for example by di Sarra et al. (2008), the spectral behaviour of the AOD in the solar spectrum largely affects the radiative efficiency and effects. This effect may be probably sorted out, for example by grouping data in different Angstrom exponent classes, and may help explaining the retrieved results and the role of desert dust.

We agree with the Reviewer on the significant role of the spectral dependence of aerosol optical properties in determining the magnitude of aerosol DRE values, as shown in earlier studies (see e.g. Hatzianastassiou et al., 2004b; 2007b; di Sarra et al., 2008). We now emphasize this point in the revised manuscript (section 1). We would like to clarify that this spectral dependence is taken into account in our study, even though we do not rely on Angstrom exponent values of aerosols, but rather consider the direct dependence by using full spectral values of AOD, ω_{aer} , and g_{aer} data. We would also like to note that by grouping data in different Angstrom exponent classes (information taken from MODIS) is not adding further information from that given in the already extended paper. Another important reason for not including analysis based on MODIS Angstrom exponent is the already reported problems with this product. For instance, Levy et al. (2010) reported that "...even when constrained to the highest confidence data, comparison of MODIS-derived Angstrom Exponent and fine AOD showed that MODIS does not provide quantitative information about aerosol size over

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land. Thus, we strongly recommend that users NOT use size products quantitatively. To avoid confusion, we plan to remove Angstrom Exponent and fine AOD parameters from future product lists...”.

- the authors show that there is a reasonable agreement between surface measurements and modelled values of downwelling shortwave irradiance (Table S1). The mean differences are however of the order of 10 W/m² or more, i.e. of the same order of the estimated radiative effects. Since the comparison is based on monthly mean values, the model data are available at a broad resolution, and the surface stations are located in areas with complex albedo, the results do not provide a direct verification of the model performance. In order to obtain a better assessment of the significance of the results, I would suggest including a comparison of the retrieved radiative effects (mainly forcing efficiencies) with those obtained in previous campaigns/measurements.

According to the results of our model validation (Figure S1 and Table S1) the model surface solar radiation fluxes agree with those from station measurements (GEBA) within 8.1 W m⁻² (or 5.3%), overall (Figure S1), and 2.2-11.7 W m⁻² (or 3-11%) at station level (Table S1). Indeed, the order of magnitude of these differences, in absolute terms, is comparable to that of model computed regional mean aerosol DRE values (-4.5 - 22.9 W m⁻², Table 1). Hence, it is difficult to assess the absolute accuracy of model DREs. Unfortunately, as explained in the first paragraph of section 3.1 (Model validation, page 7, lines 230-235), there is actually no way to directly validate the model DREs because of the unavailability of corresponding ground or reference measurements. Thus, the only way to face the problem is to ensure an improved assessment of the quality of model surface solar radiation fluxes (SSR) that are computed in the absence of aerosol particles, F_{surf,no-aerosol}, i.e. as done in the validation of our study. Such an improved assessment can be possible only through: (i) the use of surface fluxes of best quality, which is ensured by using GEBA flux data in our study, and (ii) achieving the best possible comparison/match between model and surface fluxes. With regard to the first issue, we believe that using measurements from campaigns, as suggested by the

Full Screen / Esc

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Interactive Discussion

Discussion Paper



Reviewer, would not add more to this study, given the high quality of the already used measurements. Moreover, it would even have some disadvantages, as for example limited temporal coverage due to the scarcity of available data throughout the study region and period, preventing a valuable comparison between model and measurements, especially at the monthly scale considered in this study. With regard to the second issue (ii), we agree that the different spatial resolution of model radiative fluxes, computed at 2.5degx2.5deg latitude-longitude cells, and surface measurements, referring to specific locations, can be a factor of uncertainty in our model validation, given the significant sub-grid variability of surface (for example surface albedo) and atmospheric (e.g. aerosol cloud) conditions, as indicated by the Reviewer. We agree that this uncertainty needs to be assessed. For this purpose, the spatial scales of model and surface fluxes need to be brought closer to each other. This can be done more efficiently by downscaling the model fluxes than upscaling the surface measurements. Given the nature and techniques of satellite observations and of the associated aerosol and other parameters (e.g. clouds) retrievals, to which our model computations are based and dependent, an ideal agreement between the model and surface spatial scales, is not possible. Given the characteristics of MODIS satellites, instrumentation, and inversion techniques, the best possible spatial resolution of model input data and radiative fluxes is 10 x 10 km², which corresponds to a significant increase in the spatial resolution that is actually of about 280 x 280 km². Extensive comparisons between fluxes computed at 10 x 10 km² and surface station measurements (see Benas et al., 2011) have shown that the calculated fluxes agree with surface measurements within 4.5-9.2%, comparable to the present study (3-11%), with a negative bias as also in our study (Figure S1). The similarity between the SSR computations at 10 x 10 km² and at 280 x 280 km², against surface reference measurements adds relevance to the outputs of model validation presented in this study, with regards to issues of inadequate spatial scales.

We now make reference to these findings in the revised manuscript, at the end of section 3.1.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

- the paper would benefit from a discussion of the estimated uncertainties on the retrieved forcings. For instance, section 2.2 gives an overview of the uncertainties associated with the MODIS AOD, which may be used in conjunction with the sensitivity study of section 4 to derive a first estimate of the uncertainties. Is there an estimate of the uncertainties on the used values of asymmetry parameter and single scattering albedo?

Indeed, the accuracy of model DRE values computed in this study is subject to the uncertainties of MODIS based aerosol optical properties that are used as input data to the RTM. These uncertainties have been evaluated through comparisons against ground reference measurements from AERONET stations, and were found to be equal (in absolute terms) to -2.3×10^{-4} for AOD (underestimation), -0.028 for ω_{aer} (underestimation), and -0.031 for g_{aer} (underestimation). These values were subsequently used in the derived equations relating $\Delta(\text{DRE}_i)$, where index $-i$ applies to TOA, atmosphere and surface) with ΔAOD , $\Delta\omega_{\text{aer}}$, Δg_{aer} (Fig. 7). The estimated uncertainties are equal to: (i) 0.024 , -0.009 , and -0.021 W m^{-2} , for DRE_{TOA} , DRE_{atm} , and $\text{DRE}_{\text{net-surf}}$, respectively, for AOD, (ii) -0.81 , -1.75 , and 0.51 W m^{-2} , for DRE_{TOA} , DRE_{atm} , and $\text{DRE}_{\text{net-surf}}$, respectively, for ω_{aer} , and (iii) -0.16 , 0.004 , and -0.05 W m^{-2} , for DRE_{TOA} , DRE_{atm} , and $\text{DRE}_{\text{net-surf}}$, respectively, for g_{aer} .

Reference to the above uncertainties was made in section 5, Discussion and Conclusions.

Specific comments

- p. 30016, l. 11-12: the CO₂ amount is fixed at 345 ppm. The average CO₂ in the Mediterranean in the period 2000-2007 is about 380 ppm (Artuso et al., 2009). CO₂ has a negligible influence on the SW radiative budget, but why was used such a low value?

Indeed, the carbon dioxide amount used in our study can be considered as a bit outdated, given the recent values of concentrations, as indicated by the Reviewer. No

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special care on this issue was taken just because previous sensitivity studies with our model have shown the extremely weak dependence of aerosol DREs to changes in CO₂ concentration [CO₂]. Nevertheless, as suggested by the Reviewer, we have repeated similar sensitivities, in which we increased [CO₂] from 345 to 380 ppmv. The computed new values of aerosol DRE components for [CO₂]=380 ppmv were found to differ from those for [CO₂]=345 ppmv only in the third decimal point, yielding percent differences that are smaller than 0.02%, in all cases. Therefore, we keep the already reported DRE values but make a relevant reference to this issue in section 2.3.

- p. 30019, line 22: I would add also the low ocean albedo, to which are associated the largest negative values of DRE(TOA).

Done (section 3.2.1, page 10, line 351).

- p. 30020, lines 1-6: the seasonality of DRE(TOA) does not come out clearly from figure S2. It is not possible on the basis of the plotted colours to distinguish different negative values of DRE over the sea. I would suggest using the same colour scale in the different seasons (also for figures S3 and S4), in order to allow a comparison among the different seasons. In addition, the available climatology for the central Mediterranean suggests a spring-summer maximum, and not an autumn maximum (see e.g. Moulin et al., 1998; Israelevich et al., 2002; Meloni et al., 2007; Di Iorio et al., 2009). also, the study by Papadimas et al. (2008) does not clearly show a double peak in AOD in the central Mediterranean. The double peak in spring and autumn is typical of the Greek region, dominated by Etesian winds in summer. The DRE is maximum in summer in the central Mediterranean; I believe that this is due also to the summer peak in AOD.

We agree with the Reviewer's comment, and acknowledge that the discussion in lines 1-6 had problems. The relevant part has been corrected and re-written (page 10, line 360, through to page 11, line 366).

- p. 30022, l. 20-22: the schematization that DRE(TOA) depends on aerosol scattering

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

and DRE(netsurf) on extinction is too simplistic. DRE(TOA) also depends on aerosol absorption.

According to Equation 3, DRETOA is function of reflected solar radiation at the top of atmosphere (TOA), i.e. back to space, in presence and absence of atmospheric aerosols. Given that reflected solar radiation is a function of the incoming solar radiation and the albedo (or reflectivity) of aerosols, it appears that DRETOA only depends on scattering, and not absorption, of aerosols.

- p. 30023, l. 6-9: in general, contribution of high absorbing particles from forest fires in summer is not negligible in the Mediterranean (see e.g., Pace et al., 2005; Péré et al., 2011) and may produce low single scattering albedo. It should be clarified here if the climatological values of single scattering albedo from GADS may capture interannual variations of this parameter, and which may be the effect on the retrieved radiative effects.

We agree that absorbing soot aerosol particles, which are emitted from forest fires occurring throughout Europe and the land surrounding the Black Sea during summer, as reported in the studies mentioned by the Reviewer, may imply low single scattering albedo (ω_{aer}) values. It is known that during summer a large number of forest fires occurs in Europe, and especially around the Black Sea and in the northern Balkans and also in the Iberian peninsula (van der Werf et al., 2006; hotspots/fire map products from the NASA funded Fire Information for Resource Management System, FIRMS, <http://maps.geog.umd.edu/firms/>). However, the biomass burning (forest fires) source of aerosol is less important than dust, while pollution aerosol plumes are often associated with lower AODs (Barnaba and Gobi, 2004). This has to be kept in mind that the total absorption of solar radiation by soot aerosols, is not only dependent on their single scattering albedo, which quantifies the strong absorbing ability of soot aerosols, but also on their loadings, i.e. on AOD, which are relatively low, as explained.

The absorbing ability of aerosols is quantified by ω_{aer} , while the ratio

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

DRETOA/DREnet surf describes the actual absorption of solar radiation by aerosols. Therefore, the seasonal (and inter-annual) variations of these two parameters may not be similar.

This is actually what happens. The seasonal variation of our model ω_{aer} , in terms of averaged values over the period 2000-2007, reveals a double summer minimum (in June and August), which is associated with summer biomass burning aerosols, and shows the ability of model ω_{aer} to capture the specific phenomenon. However, as seen in Fig. 2-ii, this minimum does not translate into summer minimum values of the ratio DRETOA/DREnet surf, due to the influence of the AOD seasonality as explained previously.

In order to avoid confusion related to this issue, we make a clarification in the revised text, emphasizing the difference between ω_{aer} and DRETOA/DREnet surf, as for their role for aerosol absorption.

- p. 30024, l. 12-13: the statement "On the contrary, the aerosol effects at TOA depend much less on AOD" does not seem correct. The radiative efficiency at TOA is smaller than at the surface mainly because the DRE at TOA is smaller than at the surface. The relative change in DRE, as shown by the behaviour of the ARBE, is generally larger at TOA.

We agree. We have indicated that the specific statement refers just to the absolute (and not the percentage) values of aerosol radiative effects (section 3.3, page 15, lines 500-501, and page 14, line 498).

- p. 30026, l. 16: there is a factor of 6 between the maximum and minimum monthly radiative effect both at the surface and at TOA. Thus, the seasonality is similar for both effects (although the effect at TOA is smaller).

Indeed, the factor (ratio) of summer to winter DRE values is similar at TOA and surface, but here we refer to the magnitude of seasonal differences, which is equal to 4.9 at TOA

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against 18.7 at surface. Therefore we replaced “seasonality” by “seasonal variation (or cycle)”, in section 3.4, page 16, line 565.

- p. 30033, l. 23: EARLINET stations are located on the coastal regions of the Northern Mediterranean. Few datasets provide information on the aerosol vertical distribution also inside the basin (e.g., Gobbi et al., 2000; Di Iorio et al., 2009).

Reference to these datasets has been done in the revised text.

- p. 30026-30027, section 3.5: the discussion of the interannual variations should take into account interannual variations in aerosol properties, mainly Angstrom exponent and single scattering albedo. It is not clear if interannual changes of the single scattering albedo are included (see previous comments).

As already explained, seasonal and inter-annual changes of aerosol optical properties, i.e. aerosol optical depth, single scattering albedo and asymmetry parameter, are taken into account in our model.

- p. 30032, l. 7-8: according to tables S2-S4, the largest DRE at TOA and netsurf are found in summer in the central Mediterranean.

We agree with the comment and we have corrected and re-written the relevant text, in section 5, page 21, line 734, through page 22, line 742.

- p. 30033, l. 1-3: the effects on the temperature profile, and consequently on the vertical stability, cloud processes, etc., critically depend on the aerosol vertical distribution. In many cases the dust absorbing particles travel above the boundary layer, and the associated heating takes place in specific altitude ranges. Thus, the temperature lapse rate may increase in some vertical intervals, affecting in a more complex way atmospheric stability and cloud processes.

Reference to the vertical dimension of aerosol induced changes of temperature lapse rates, atmospheric stability etc, is made in the revised text, section 5.

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- p. 30033, l. 14: not necessarily "benefits". I would suggest "effects".

We replaced "benefits" by "consequences" (page 23, line 776).

- p. 30033, l. 28: as far as I know, CALIPSO provides backscatter vertical profiles, not only AOD.

We agree, but we meant that not all three model input aerosol optical properties, AOD, ω_{aer} , and g_{aer} , are provided by CALIOP/CALIPSO. The relevant text has been modified accordingly.

- Figure 3: please, clarify if the plotted values are monthly or annual averages. Part of the large spread may be due to varying illumination conditions (latitude + time), in addition to changing aerosol properties and surface albedo.

We have indicated in the caption of Figure 3 that monthly mean values of DRE and AOD values are used in the scatterplots.

Technical corrections

- the papers by Hatzianastassiou et al. (2007) is often cited as (2007a). See e.g. p. 30013, line 20; p. 30015 l. 21, etc.

The paper has been corrected accordingly.

- p. 30024, l. 2-3: DRE, not E(AOD), is plotted versus AOD to derive the forcing efficiency.

The text has been corrected accordingly (section 3.3).

- p. 30031, l. 6: have been used.

Done (section 5).

References

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/11/C15593/2012/acpd-11-C15593-2012-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 30009, 2011.

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