Aerosol radiative forcing during African desert dust events (2005-2010) over south-eastern Spain.

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Answers to Reviewer #1 comments:

The authors greatly acknowledge the anonymous Reviewer for carefully reading the manuscript and providing constructive comments that will lead to an improved paper. Following referee's suggestions we have made some changes in the revised manuscript. Here we included after each one of the reviewer's comment our responses in bold.

General comments

This paper deals with African dust direct radiative forcing estimation that relies on aerosol optical characteristics retrieved from principal plane photometric measurements. The presented study utilizes observations at a site in south-eastern Spain and focuses on long-term observations of dust transported from different sources in the northern Africa. The authors report high aerosol absorption that leads to stronger, relative to other studies, radiative forcing. In general, the defined in the paper objectives are important. Particular interest presents usage of principal plane photometer measurements for aerosol optical characterization. At the same time, I have serious concerns regarding poor description of the methodological part. In addition, the paper presents only values of forcing and does not show the actual retrieved aerosol parameters. Instead of providing direct discussion of the methodology and retrieved aerosol properties, the authors provide only references to previous publications. This forces a reader to search in literature for information essential for proper understanding of what is done in this study. Moreover, even after looking the referred literature it is difficult to find consistency (although it can exist) between presented here forcings and reported in other two papers aerosol properties. In addition, in the referred "methodology paper" I found only about one page long description of the aerosol retrieval algorithm that did not contained the details needed to address all my questions. As a result the paper leaves impression of a non-transparent study, especially because the authors suggest that the main originality of this study is in using their own aerosol retrieval products instead of using the standard retrieval products provided by AERONET. In addition, the authors tend to make conclusions about "excellent agreements" without providing convincing and balanced discussion required for justification of such conclusion. Without proper justification the usage of such terms as "excellent agreements" in the text gives impression of immaturity of the study or its presentation.

In summary, I have concluded that the paper is not acceptable for publication in ACP in its present state and some significant improvements should be done in order to make paper acceptable. The corrections should address my general comments above and the specific remarks below.

According to the referee's suggestions we have provided a more detailed description of methodology used in this study with comprehensive sensitivity test and assessment of uncertainties in the revised manuscript (see our responses to the specific comments below and the Answers to Editor). Also, we have included in the new version of the manuscript the aerosol optical and microphysical properties retrieved by this method during desert dust events. On the other hand, we have moderated the conclusions as "excellent agreements" in the new text. In order to justify the agreements between experimental and simulated results we have computed the relative differences between the experimental and modeled irradiance values. The analysis shows that the radiative transfer model slightly overestimates (mean bias of 3%) the experimental global irradiances. This information will be included in the revised manuscript.

1) Abstract, line 9: You wrote, "The SBDART modeled global irradiances at surface have been successfully validated against experimental measurements obtained by CM-11 pyranometer, indicating the reliability of the radiative transfer model used in this work for the ARF calculations." You can be more specific and provide the estimated errors.

As we commented before we have computed the relative differences between the experimental and modeled irradiance values. The mean relative difference was 3%. This information will be included in the revised manuscript.

2) Abstract, line 13: The values of aerosol radiative forcing are provided, but it is not clear either they are averages of instantaneous values or 24 hours average?

In the first version of the manuscript, we have derived the mean aerosol radiative forcing during desert dust events from the instantaneous values. However, in the revised version, according to reviewers' suggestions, we have computed the daily mean aerosol radiative forcing (24 hours averages). In addition, we have recalculated the mean forcing for the different desert dust origin sectors as well as the monthly mean forcing from daily mean forcing values. These results will be presented and discussed in the revised manuscript.

3) Abstract, line 18: You wrote, "These results suggest that the African dust caused local atmospheric heating over the study location." This obvious conclusion is not necessary to report in the abstract. If you want to write something about atmospheric heating please provide a value, e.g. x K/day.

Following the reviewer's suggestions we computed daily mean heating rate values. The mean heating rate value (derived from daily mean values) for desert dust events from 2005 to 2010 at Granada was 1.5 K/day. This information will be included in the revised manuscript.

4) Abstract, line 23: Do you provide instantaneous values or an average of these values? If they are instantaneous, then for what SZA? These numbers do not mean much without understanding of how it was calculated.

In the old version of the manuscript the provided mean aerosol forcing values were calculated from instantaneous values. However, according to reviewer's suggestions we have recalculated these mean aerosol forcing values from daily mean aerosol forcing values (24 hours averages). These results will be presented in the revised manuscript.

5) Abstract, line 23: In abstract you report some results for sectors A, B, C. It is not possible to know what these sectors are without reading the paper. Abstract should be self-sufficient.

In order to clarify this, we have included the areas that are within each sector in the abstract of the revised manuscript.

6) Abstract, line 24: You claim that the found forcing values are larger than many of values found in literature due to the presence of more absorbing atmospheric particles during African desert dust intrusions over the study area. Maybe it is simply because of not consistent comparison with values in literature? Maybe literature gives forcing per 24 hours (daily averages), while you give an average during daylight duration only? Maybe solar geometry is different? It does not seem to me that SSA values reported in (Valenzuela, 2012a) are much lower than in many other studies of African dust. Otherwise, please be more specific and clear.

As we have commented before, in the new version of the manuscript we include daily mean aerosol radiative forcing values (24 hours averages). In order to be more consistent in our comparison with other studies, we made some changes in old Table 3 (Table 5 in new version of the manuscript). In this sense, we have included additional information about each study in this table (e.g. the wavelength range where the radiative forcing was calculated and the used surface albedo) and we only included studies that reported daily mean aerosol radiative forcing (24 hours averages) during desert dust episodes. The results of the comparisons showed small differences between our computed aerosol radiative forcing and those reported by other authors during desert events. These differences may likely be related to the differences in the methods used, measurement periods, desert dust load, and chemical composition of dust, aerosol mixing state and surface albedo. All this information will be included in the revised manuscript and new Table 5 (Table 1 enclosed below).

Respect to the comment "SSA values reported in (Valenzuela, 2012a) are much lower than in many other studies of African dust", we refer that the values of single scattering albedo obtained during desert dust events reported in Valenzuela et al. (2012a) are much lower than those given for "pure desert dust" (e.g. Dubovik et al. 2002b). However, single scattering albedo values reported by Valenzuela et al. (2012a) are comparable to those obtained in other urban Mediterranean sites (e.g. Tafuro et al., 2006). We have clarified this point in the new version of the manuscript. 7) Introduction, p. 6595, line 17: "region" is used twice, please reword.

We have replaced the word ``*region''* by the word ``*layer''*.

8) Introduction, p. 6597-6598: You describe advantages of aerosol retrieval from principal plane observations, but you do not mention at all any limitation. There is not discussion of fitting errors, quality controls, etc.

One of the major difficulties of the retrieval of the sky radiance in principal plane and almucantar configurations is the cloud screening procedure. This quality assurance is more difficult, and possibly may have more errors, than for almucantar configuration, where symmetry analysis of the left and right parts of the sky radiance measurements can be used. However, the symmetry criteria cannot be applied on the principal plane data. In this study, to eliminate cloud contaminated aerosol optical depths, and the corresponding principal plane sky radiance measurements, first we applied the triplet stability criteria (Smirnov et al., 2000) to the spectral extinction data measured at the beginning of the principal plane scan. In addition, to consider the possible influence of thin clouds and spatially inhomogeneous clouds, which are difficult to completely eliminate and could contribute to the sky radiance errors, we have smoothed the principal plane sky radiances by a moving average smoothing algorithm with slide window of fivepoint width. If the number of scattering angles in the measured sky radiance distributions is less than 20, the measurements record is eliminated. In addition, we only consider as out parameters of the inversion code the aerosol properties retrieved with fitting accuracy (relative differences between the measured and computed normalized principal plane sky radiances) less than 10%, and also RMSE -root mean square error- less than 5% for aerosol optical depth. Finally, to eliminate cloud contaminated principal plane sky radiance observations that passed the previous filters we used a manual inspection of an All-Sky Imager (e.g. Cazorla et al., 2008). In this sense, the methodology is not completely automatic (see Editor Responses).

9) Introduction, p. 6598, line 5: Something is missing in this sentence.

Thank you for your comment. This mistake has been corrected.

10) Instrumentation part, p. 6599, line 11: Here you report the pyranometer spectral range of 310 - 3200 nm. In abstract you report 300 - 2800 nm for global irradiance simulations. Please check. If spectral ranges of pyranometer and of simulations are different, please provide an estimate of uncertainty due to this difference.

The given spectral range of the pyranometer in old manuscript version was incorrect. The correct spectral range is 310-2800 nm. The global irradiance simulations were done in 310 - 2800 nm spectral range. Therefore, there were no differences in the spectral range of pyranometer and of simulations. We have corrected these typographical errors in the new version of the manuscript.

11) Instrumentation part, p. 6599, line 13: You wrote, "the calibration factor stability has been periodically checked against a reference CM-11 pyranometer". So, what are results of these checks? How often it was done? The data consists about 5 years. Was it necessary to apply any corrections during this period? Also, as I understand the global irradiance was measured by not shadowed pyranometer. In this case there can be an increase of uncertainty during morning and afternoon due to non-uniform illumination of the detector by the direct solar beam. How significant this uncertainty is? Further on in the paper you report smaller that 4 % difference between simulated and measured irradiances, which is similar or even less than expected accuracy of the measurements. In Figure 1 I see that the differences can be about 10 %. I would suggest showing absolute or relative errors, not a correlation scatter plot where tens Wm-2 errors are not distinguishable on hundreds Wm-2 scale. For example you can show relative errors vs. absolute values of global irradiance.

The stability of the calibration factor has been periodically checked using a reference CM-11 instrument in four inter-comparison campaigns performed between March 2005 and June 2010. The results derived from these inter-comparison campaigns showed relative variations of the calibrations coefficients smaller than 1% for the 2005-2010 period which guarantees the stability of the calibration factor of the instrument used in this study. We think that, due to this great stability, no corrections are necessary during the period of study. This information will be included in the new version of the manuscript.

Regarding the uncertainty associated with the non-uniform illumination of the CM-11 detector by the direct solar beam, we think that this effect could be relevant for high solar zenith angles (SZA). In order to avoid this effect, in the comparison between simulated and CM-11 measured irradiances we have only used experimental global irradiance values recorded for SZA smaller than 75°. Therefore, in our opinion, the influence of this effect on the differences between simulated and measured irradiances is negligible.

We agree with the reviewer in that a plot showing the relative errors against the absolute values of global irradiance would be very explanatory. According to the reviewer's suggestion, this new figure is shown below as Figure 1 and it will be included in the revised version of the paper as Figure 3.

12) Methodology and data, p. 6600, line 4-7: Analysis in the current paper heavily relies on aerosol characteristics retrieved from principal plane photometer measurements. The authors state that the method has been previously described by (Olmo et al., 2008). However, Olmo et al., 2008 state in the abstract that "In this paper we test a parameterization of the particle shape in size distribution, single-scattering albedo, phase function and asymmetry parameter retrieval from beam and sky-radiance measurements, based on the model Skyrad.pack, taking into account the principal plane measurements configuration". After looking through the paper I agree with the statement in the abstract that this paper is rather a test very few cases examined. But, it is not enough only to cite this paper without providing in deep discussion of methodology with comprehensive sensitivity tests and assessment of uncertainties. In any case, the paper by Olmo et al. (2008) is a limited study that cannot serve as a reference paper containing comprehensive description of the retrieval methodology. For example, you say that your method is an improvement of Nakajima et al. (1996) code where the most important change is the substitution of spherical model used in the original algorithm by spheroid model. However, Skyrad described in Nakajima et al. (1996) does not retrieve complex refractive index. How the complex refractive index retrieval is treated in your work? Please describe clearly your methodology of aerosol retrieval before making conclusions about radiative forcing.

Detailed description of the algorithm used in this work has been summarized in the new version of the manuscript, including sensitivity tests and assessment of the uncertainties. Please, see information about this question in our response to the Editor.

13) Methodology and data, p. 6600, line 9-15: How well the retrieved spectral aerosol optical properties represent the whole broadband spectral range? Are they only for nominal CIMEL photometer wavelengths? What is assumed for other wavelengths in broadband spectrum?

The global irradiances have been obtained with SBDART radiative transfer model using as input in the experimental aerosol information (spectral aerosol optical depth, spectral single scattering albedo and spectral asymmetry parameter) derived from the principal plane retrievals. Logarithmic interpolation among the available spectral values (extrapolation for $\lambda < 414$ nm or $\lambda > 860$ nm) was used to supply SBDART with aerosol optical depths covering the entire wavelength range of the calculation (310–2800 nm). Linear extrapolation/interpolation is used for spectral single scattering albedo and spectral asymmetry parameter.

14) Methodology and data, p. 6600, line 17: You say that surface albedo is fixed to 0.15. At what wavelength? What is spectral dependence? I guess you account for spectral dependence of surface albedo; otherwise your calculations of forcing at top of the atmosphere are highly uncertain.

In the old version of the manuscript we used a fixed value of 0.15 for surface albedo in the simulations. However, according to referee's suggestion we have recalculated all simulated values of the aerosol radiative forcing taking into account the spectral dependence of the surface albedo at the study site. For that, we used as input in the SBDART code the averages surface spectral albedo provided by the AERONET algorithm which is based on a dynamic spectral and spatial model estimation at four wavelengths: 0.05 at 440 nm, 0.16 at 675 nm, 0.31 at 870 nm and 0.32 at 1020 nm. This algorithm adopted the Lie-Ross model for land surface covers (Lucht and Roujean, 2000), considering the bidirectional reflectance distributions taken from MODIS (Moody et al., 2005). All this new information and obtained results have been added to the revised version of the manuscript.

15) Results and discussion, p. 6603, line 18: Interpretation of provided here forcing values is not clear. What is "overall mean"? As far as I understand, these are not 24

hours averages. Why you conclude that these values indicate "significant Earthatmosphere cooling" and "significant atmospheric warming"? Can you provide corresponding values of heating rate or compare them to the values for some other situation?

With "overall mean" we refer to the average of mean daily aerosol radiative forcing for desert dust events observed during the entire period. As we have commented before, we have recomputed the daily mean aerosol radiative forcing (24 hours averages) during desert dust events. In addition, to justify the atmospheric heating or cooling we have computed the daily mean heating rate. For instance, the daily mean heating rate (k/day) above surface was 1.6 K/day, 1.5 K/day and 1.4 K/day when air masses were transported from sector A (North Morocco; Northwest Algeria), sector B (Western Sahara, Northwest Mauritania and Southwest Algeria) and sector C (Eastern Algeria, Tunisia), respectively. The daily ARF values according to the classification by desert dust sectors origin are shown in new Table 5. This information will be included in new version of the manuscript.

16) Results and discussion, p. 6606, line 6: Why in Fig. 4 you analyze forcing vs. AOD for the SZA lower than 65 degree? Please explain this choice. Can be that a large "cloud" of points in Fig. 4 for TOA is because you take forcings for different SZAs (range 65 to 80 degree) and this is why the correlation is poor. In other words, you have dependence not only on AOD, but also on SZA and therefore the derived forcing efficiency is very uncertain. It seems that correlation for surface forcing vs. AOD is better, which is because of weaker dependence of the forcing on SZA. By the way, why the offsets are removed in the linear regression equations of Fig. 4? Are they equal to zero? How the provided in the text uncertainties of the forcing efficiency were calculated?

In old version of the manuscript we have computed the instantaneous aerosol radiative forcing as function of the solar zenith angle (figure 3 in old version). As can be seen from this figure there is a strong change in aerosol radiative forcing at solar zenith angle around 65-70 degrees. For this reason, we have used aerosol forcing at surface and TOA for zenith angles below 65 degrees in figure 4. So we think that the poor correlation for TOA forcing vs. AOD is not related with solar zenith angles. Nevertheless, we have removed the figure 4 from the new version of the manuscript. Furthermore, we have computed daily aerosol radiative forcing efficiency (ARFE) as the ratio of daily aerosol radiative forcing by the corresponding daily mean AOD (440 nm). Using these daily ARFE we also computed the ARFE for each desert dust sector origin. The new ARFE results were included in Table 5 in the new version of the manuscript.

17) Results and discussion, p. 6607, line 10-: Your forcing simulations are for spectral range 300 - 2800 or 3200 nm. Is it the same range as used in AERONET forcing calculations? If not, please check and discuss the influence on the conclusions about the comparison.

AERONET forcing calculations are done in 0.2-4.0 μ m spectral ranges. However, the AERONET procedure used to compute the aerosol radiative forcing at surface

is different to our method at surface. In AERONET the aerosol radiative forcing at surface is computed as;

 $\Delta F_{surface} = F^{\downarrow A}_{surface} - F^{\downarrow c}_{surface}$

where $F^{\downarrow A}_{surface}$ indicate the downward global irradiances at surface with aerosol presence and $F^{\downarrow c}_{surface}$ indicate the downward global irradiances at surface without aerosol presence.

Therefore, we can not directly compare our aerosol radiative forcing at surface to those given by AERONET. However, we can compare the instantaneous global irradiances simulated with SBDART model and the corresponding instantaneous global irradiances provided by AERONET. So, for comparing with AERONET we have run the SBDART model in the same spectral range as used in AERONET. The analysis shows that the relative differences between upwelling global irradiances at TOA and down welling global irradiances at surface simulated with SBDART model and the provided by AERONET are 0.8% and 2.4%, respectively. Figure 2 shown in the bottom of this report (Figure 4 in new version of the manuscript) displays the scatter plots of the instantaneous global irradiances using SBDART model against corresponding AERONET fluxes for a) downward fluxes at surface and b) upward fluxes at TOA. The black lines are the linear fits, with the equations regression and determination coefficients and biases. The results of this comparison will be included in the revised manuscript.

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Table 1: Daily aerosol radiative forcing (W/m²) and daily aerosol radiative forcing efficiency (W/m² per unit of $AOD(\lambda)$) at surface, TOA and in the atmosphere observed over different locations during desert dust events. The second column (λ) indicates the spectral range considered and third column shows the surface albedo (α) used in each study.

Reference	$\lambda(\mu m)$	α	DARF TOA	DARF Surface	DARF Atmosphere	DARFE TOA	DARFE Surface	Location
Meloni et al. (2005) [1]	0.4 - 0.7	0.02-0.37	-5.1 to -8.7	-11.0 to -14.2	3.7 to 9	-15.0 to -16.4	-28.4 to -30.1	Lampedusa, Italy
Derimian et al.(2006) [2]] 0.175 - 2.270	0.23-0.35	-2.1	-6.4		-22	-65	Negev, Israel
Derimian et al.(2008) [3]] 0.2 – 4.0	spectral depen.	- 8.1	-29.1	21.0	-15.7	-56.4	M'Bour, Senegal
Prasad et al.(2007) [4]	0.3 – 3.0	0.25	- 2.9 to -26	-29.5 to -87.5		-17±3	- 46±3	Kanpur, India
Lyamani et al. (2006) [5]] 0.4 – 0.7	0.15	-4.0	-20.4	16.4	-14.5	-73.4	Granada, Spain
Di Sarra et al. (2011) [6]	0.3 - 3.0	0.07		-69.9±3.4			-59.9±2.6	Lampedusa, Italy
Huang et al. (2009) [7]	0.175 - 4.0	spectral depen.	14.11	-64.72	78.8			Taklimakan Desert China
Saha et al.(2008) [8]	0.28 - 2.8	spectral depen.	7.7 to -9.8	-61.8 to -64.4	54.1 to 54.6	-9.7 to -12.4	- 78.2 to -81.5	Toulon, France
Present study	0.31 - 2.8	spectral depen.	-5±5	-20±12	15±9	-17±7	-74±12	Granada, Spain
Present study	0.31 - 2.8	spectral depen.	-7±5	-21±9	14±7	-20±9	-70±14	Granada, Spain
Present study	0.31 - 2.8	spectral depen.	-6±5	-18±9	12±8	-22±10	-65±16	Granada, Spain

Method: [1] Surface albedo varies between 0.02 at 20° and 0.37 at 90° SZA. Unit for DARF is W m⁻² AOD⁻¹ (500 nm). [2] Mixture of desert dust and anthropogenic aerosol. Unit for DARF is W m⁻² AOD⁻¹ (550 nm). [3] Takes into account the non-sphericity of dust particles for simulating radiative effects. Unit for DARF is W m⁻² AOD⁻¹ (440 nm). [4] Unit for DARF is W m⁻² AOD⁻¹ (500 nm). [5] Fixed surface albedo of 0.15. Unit for DARF is W m⁻² AOD⁻¹ (675 nm). [6] The surface albedo has been calculated as the weighted average of land and ocean albedo over a 10 Km diameter area around the measurement site. [7] Takes into account the vertical distributions of the dust aerosol extinction coefficient. [8] Unit for DARF is W m⁻² AOD⁻¹ (440 nm).

Figure 1: Relative differences between modeled (SBDART) and experimental (CM-11) downward irradiances against experimental values.



Figure 2: Scatter plots of the instantaneous global irradiances using SBDART model against corresponding AERONET fluxes for a) downward fluxes at surface and b) upward fluxes at TOA. The black lines are the linear fits, with the equations regression and correlation coefficients and biases.



