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

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Answers to Reviewer #2 comments:

We would like to thank the reviewer for the helpful comments. 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

The paper aims at characterizing the aerosol radiative forcing and forcing efficiency due to desert dust that at a Spanish location (Granada) located relatively close to the Mediterranean Sea and in the path of dust-loaded air masses originating in the North African desert areas. Although the topic is sound, the methodology is poorly explained and the paper lacks a rigorous analysis in many senses. The paper would need substantial revision before it could be considered for publication in ACP and I suggest it is rejected and encourage authors to re-submit once they re-write the manuscript and address the concerns that follow below.

We appreciate the reviewer's comments. According to referee's suggestions we have made important changes in the revised manuscript. In this sense, we have added in the Methodology section a more detailed description of the non-spherical inversion method used in this study (see our specific response below). In addition, we have added a comprehensive sensitivity test and assessment of uncertainties in the retrieved aerosol properties in the revised manuscript. Please find the detailed description of the algorithm used in this work, including sensitivity tests and assessment of the uncertainties in our response to the Editor. Furthermore, we have included in revised manuscript a detailed description of the method used to guarantee the quality of data presented in this study. In order to provide a more rigorous analysis of data we have applied different statistical tests to the data. The discussion of these results was incorporated in the new version of the manuscript.

1. The result section and the conclusions are full of vague statements (such as 'show agreement', 'high degree of agreement' which are not demonstrated at all. A real quantification is needed to support the results. Of special interest are the differences in ARF and ARFE with respect to the AERONET retrievals, which are not quantified. The same applies to the comparison between model and flux measurements. There are also vague statements and even speculations without observational base ('…could be due

to…'). The authors should stick to the observed data and avoid speculations, for instance about the SSA.

In order to justify our statements about the agreement between data, we have applied statistical test to the different data and we have computed the relative differences between the different data set (please read the response to the Editor). The results of these analyses will be included and discussed in the new version of the manuscript. The AERONET procedure used to compute the aerosol radiative forcing at surface is different to the used in this study. Thus, 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 (0.2-4.0 μm) as used in AERONET. A statistical analysis of this comparison will be included in the new version of the manuscript. In order to quantify the difference between model and flux measurements we computed the relative differences between the experimental and modeled irradiance values. The mean relative difference was about 3%. More detailed information about this point will be included in the revised manuscript. In order to better justify our retrievals we have included in the revised manuscript information about the optical and microphysical properties.

2. The main body of the paper should be consistent and adequately justified with the data that are presented. Citations to Valenzuela et al. (2012a,b) are not enough and the authors should consider including information about AOT and SSA at the site. You mention the seasonal AOT both in the abstract and the conclusions, but this not given in the paper! The number of investigated days (apart from or instead of the number of observations) should be given too.

We will include in the revised manuscript a summary table with complete information about the optical and microphysical properties which also will include the number of investigated days (Table 1 enclosed below).

Specific comments:

1. It is not clear to me whether all data or only data during dust episodes are considered in the analysis. Note the sentence: 'In fact, the monthly AOD (440 nm) value was slightly larger in June (0.37) than in July (0.31).' and the sentence 'the largest values of ARF at surface in April and July coincided with the lowest monthly values of the single scattering albedo for those months'. If only data from dust events is considered, the analysis is poor and needs to be enhanced with the aerosol properties in the absence of dust. If the complete dataset is considered (this is not clear to me), such AOT and SSA should be shown (see general comment 2).

In this study we only used data obtained during desert dust intrusions. We clarify this information in the revised manuscript. In addition, we have added to figure 5 the monthly mean values of $AOD(\lambda)$ and $\omega_0(\lambda)$ obtained during dust intrusions. Our study focuses on the analysis of aerosol radiative forcing during desert dust

intrusions from different origin sectors. So, we think that the analysis of aerosol properties in the absence of dust is beyond the scope of this study.

2. There is also need for more rigorous statistical analysis. The ARF differences among sectors might not be significant. Please use any of the well-known specific tools (significance tests) to analyze whether the three populations of data are different in a statistical sense. At a certain confidence level, are the three ARF subsets statistically distinguishable? Note that the measurement uncertainty plays a role at this point too.

According to referee's suggestions we have applied a Kolmogorov-non-parametric test to the three ARF subsets. The test revealed that ARF at TOA for sector A (North Morocco; Northwest Algeria) was significantly different from the others two sectors. In addition, the test showed that ARF at TOA was not significantly different between sectors B (Western Sahara, Northwest Mauritania and Southwest Algeria) and C (Eastern Algeria, Tunisia). However, there were no significant differences in ARF at surface between the different origin sectors (Table 2 enclosed below). These results will be included in a new table in the revised manuscript.

3. How did the authors accomplish the data quality assurance in principal plane re-trievals? Neither the cloud screening nor the error estimation are described. Compared to Olmo et al (2008) this paper deals with a larger dataset, so a manual approach does not seem adequate. What is the uncertainty in both the SSA and the ARF? Do you apply any limitation (in AOT or other parameters) as it is done in AERONET level 2.0 database? Neither method nor result is shown that assures the quality of your retrievals.

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 five-point width. If the number of scattering angles in the measured sky radiance distributions is less than 20, the measurements record is eliminated. Also, we only consider as output parameters of the inversion code the aerosol properties retrieved with fitting accuracy (relative differences between the measured and computed 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.

For aerosol optical depth higher than 0.4, uncertainty in single scattering albedo is about 0.03. However, for aerosol optical depth < 0.2, this uncertainty falls to 0.07. In our study, we have only used the retrieved single scattering albedo obtained for aerosol optical depth values higher than 0.2. On the other hand, uncertainty in ARF computed using SBDART model is related to uncertainties in the input aerosol optical parameter and in the spectral surface albedo as well as meteorological parameters considered by radiative transfer model. The overall uncertainty in the estimated ARF due to deviations in simulation is about ~10–15% (Prasad et al., 2007; Alam et al., 2012). This information has been clarified and included in the new version of the manuscript. More details about methodology utilized in this work and quality of retrievals can be seen in our response to the Editor.

4. Have you tried using the CM11 flux measurements for evaluating radiative forcing at surface? Seems to be straightforward and this reviewer would find interesting to see the differences with the 3-step methodology (acquisition of sky radiances with a Cimel, inversion and finally flux modeling).

In our paper, the surface aerosol radiative forcing (ARF) has been derived from the following expression:

$$ARF = \left(F_d - F_u\right) - \left(F_d^0 - F_u^0\right) \quad \textbf{(1)}$$

where F_d is the downwelling global solar irradiance and F_u is the upwelling surface flux under cloud-free conditions and superscript "0" denotes fluxes in absence of atmospheric aerosol. The four fluxes were simulated by the SBDART code.

The CM-11 pyranometer located at the study site only measures the downwelling global solar irradiance. Thus, it is not possible to obtain experimental values of the net flux under cloud-free conditions (left term of equation 1). Therefore, we cannot evaluate the radiative forcing at surface using the CM-11 flux measurements by means of the expression 1.

Several authors have used a fixed surface albedo (α) to relate the downwelling and upwelling fluxes as:

$$F_u = \alpha \cdot F_d \quad \textbf{(2)}$$

Thus, combining equations (1) and (2), the ARF can be expressed as a function of the downwelling flux and the surface albedo:

$$ARF = (1-\alpha)\left[F_d - F_d^0\right] \quad \textbf{(3)}$$

This last equation allows to determinate the ARF from the experimental data recorded by our CM-11 instrument (F_d) and simulated values for aerosol-free conditions (F_d^0).

Although it could be interesting to compare the modelled ARF values inferred from equation 1 with respect to those ARF values derived from the semi-empirical

equation 3 using the CM11 flux measurements, considering the extension of the manuscript after the review process we think that this subject could be addressed in a future paper.

5. Please give information on how you calculate monthly ARF. This is important in order to allow comparison with other publications. Have you considered calculating monthly ARF with and without dust events, to estimate the contribution in ARF of dust with respect to the dust-free conditions?

In the first version of the manuscript, we have computed the monthly mean values of the aerosol radiative forcing using instantaneous values. However, according to the reviewers' suggestions we have recalculated the daily mean values of aerosol radiative forcing (24 hours averages). In addition, we have computed the monthly mean values of the aerosol radiative forcing from those daily mean aerosol radiative forcing values. This information will be included in the revised manuscript. On other hand, the aim the present work was the evaluation of aerosol radiative forcing during desert dust episodes and hence the analysis of ARF without dust events is beyond the scope of our study.

6. If (as stated by the authors) the information provided in table 3 may not be comparable, the table is nonsense to me. This is also linked to the comment above. The method how you calculate forcing must be provided.

In order to be more consistent in our comparison with other studies, we made some changes in this table (Table 3 enclosed below). In this sense, we have extended the 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 have only included studies that reported daily mean aerosol radiative forcing (24 hours averages) during desert dust episodes.

7. Abstract: if AERONET is used as a well-established reference to prove the goodness of the analysis, quantitative differences should be given (see general comment 1).

As we commented before the ARF obtained in this study are not directly comparable with those provided by AERONET. Thus, we compared the instantaneous global irradiances simulated with SBDART model and the corresponding values provided by AERONET. For this, we have run the SBDART model in the same spectral range (0.2-4.0 μm) used by AERONET. The analysis shows that the relative differences between upwelling global irradiances at TOA and downwelling global irradiances at surface simulated with SBDART model and the provided by AERONET are of 0.8% and 2.4%, respectively. Thus, we have included a new plot in the manuscript (Figure 1 enclosed below) which shows the scatter plots of the instantaneous global irradiances using SBDART model against corresponding AERONET fluxes. These results will be including in the revised manuscript.

8. There are abbreviations (e.g. 'ARFSurface') that are used only once in the text. Please be consistent.

Thank you for your comment. This expression has been changed.

9. If the forcing efficiency is (supposedly) seasonal dependent, why are all available data mixed up together in Figure 4?

In the old version of the manuscript we have applied a simple method (linear regression between DARF and AOD) for calculating the aerosol radiative forcing efficiency, ARFE. However, according to referees suggestion we have computed the 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. In addition we have removed the figure 4 from the new version of the manuscript.

Minor comments

- The English usage must be improved. I recommend that a native speaker edits the text.
- The sentence ‘This result suggests a relevant absorption of solar radiation in the atmosphere, leading to significant atmospheric warming.’ is nonsense given that such data (atmospheric ARF) are available. Please rephrase. - Fig. 4: the regression statistics are missing (correlation coefficients, etc.)

The revised manuscript will be improved in English language. According to referee’s suggestion we rephrased the sentence ‘This result suggests a relevant absorption of solar radiation in the atmosphere, leading to significant atmospheric warming.’ We have removed Figure 4 from the text due to the change made in the new version of the manuscript

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Table 1: The number of desert dust days, number of measurements recorded by sun-photometer and the daily mean $AOD(\lambda)$, $\omega(\lambda)$ and $g(\lambda)$ values.

	Sector A	Sector B	Sector C
Days	86	56	41
Measurements	426	287	195
$AOD(440nm)$	0.28±0.18	0.30±0.13	0.28±0.13
$\omega_0(440nm)$	0.89±0.03	0.89±0.03	0.90±0.03
$\omega_0(1020nm)$	0.90±0.03	0.92±0.03	0.92±0.03
$g(440nm)$	0.69±0.01	0.70±0.01	0.68±0.01
$g(1020nm)$	0.67±0.01	0.67±0.01	0.67±0.01

Table 2: The p values of the Kolmogorov-Smirnov non-parametric test for each pair of origin sectors, with *ARF* at TOA tests above the diagonal and *ARF* at surface tests below it. Values p< 0.05 indicates statistical significant differences between means at the 95% confidence level.

	Sector A	Sector B	Sector C
	ARF at TOA		
Sector A	---------	**0.008**	**0.009**
Sector B	0.493	--------	0.601
Sector C	0.555	0.084	---------
	ARF at surface		

Table 3: Daily aerosol radiative forcing (W/m^2) and daily aerosol radiative forcing efficiency (W/m^2 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	λ(µm)	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 20º and 0.37 at 90º 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: 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.