Dear Editor,

please find below the answers to the remarks of the reviewes. We want to thank to the reviewers for their constructive comments.

Our replies are highlighted with '++' symbols and red font.

The authors

<u>Referee 1</u>

General comments:

Mateos et al., 2014 presents aerosol radiative effect (ARE) obtained at six Iberian sites in different period for each station covering from 2000 to 2012. Also, the aerosol forcing efficiency (AFE) is obtained. The ARE and AFE were calculated in the UV, VIS, NIR, and SW spectral regions. The last is a novel result for the region. It is used data on aerosol optical properties from AERONET and atmospheric radiative transfer models. So, the paper addresses relevant scientific questions within the scope of ACP. The principal objective of the paper is to analyze the behavior or trend in aerosol optical properties and its radiative effect to produce a characterization of the aerosol over the Iberian Peninsula. There were also obtained the relation of the ARE with the aerosols properties. This paper is interesting, but I have some comments that are informed below. I have two points to specify, the first related with the discussion of the results and a description of the results or phrases. Some examples are given below in the specific comments. The second point is related with the presentation of data you should give more details in the discussions, when comparing with other reports or show dataset. The originality of the results must be showed. I propose to approve the paper for publication, after some major revisions, corrections and modifications. I hope the future version will have an improvement of the quality of the results.

Specific comments:

Abstract; Page 8781 Line 7: "climatology"? I suggest substituting by "climatological or climatologic".

++ The sentence was rephrased.

Introduction

Page 8783 Line 15: Which six stations? You used the term "the six stations" and It is the first time you mention these stations in the text: : : Which time period? It is the first time mentioned in the text. I propose to organize this paragraph, introducing these points at least in general sense.

++ The sentence was rephrased including the name of the six sites and time periods: "Hence, aerosol climatology at six stations (Palencia, Barcelona, Cabo da Roca, Évora, Granada, and El Arenosillo) is also carried out for different time periods between 2001 and 2012. "

Section 2

Page 8784 Line 1: My suggestion is to change the title "Aerosol ground-based data". The term ground-based is usually applied to measurements or instruments.

++ The new title is "2. Columnar aerosol optical data". We added columnar to better distinguish with the aerosol "in-situ" measurements.

Page 8784 Line 4:

Why do you select only these six stations? Did you based in a specific criterion, the number of years? I propose to add this information in the text.

++ Yes, the six longest sites in the AERONET network (and level 2.0 data) are used in the analysis. We added this information to the manuscript.

Page 8784 Line 4: Cimel CE-318 ??

++ Yes, the model was indicated in the text.

Page 8784 Line 7: Which selected wavelengths? Please, could you mention?

++ We indicated that the CIMEL wavelengths are in the range 340-1020nm. Few lines after, the reviewer can read the four wavelengths used in this work.

Page 8784 Line 24: Why do you mention "absolute uncertainty about 0.03-0.05"? Dubovik et al., 2000 report in Table 4 as higher limit the value of 0.07.

++ Thank you, it was a typo which has been corrected.

Page

8785 Line 10: Why do you select the value 0.15 as a threshold to AOD440? Please, explain in the text. Was the value of AOD = 0.15 excluded in the considerations? Because do you show the sign higher than or below than but not equal.

++ The choice of AOD = 0.15 is arbitrary but it is motivated to obtain reliable data together with the longest dataset as possible. Furthermore, the 0.15 usually constitutes a threshold to identify clean conditions (low turbidity), and the impact of aerosol properties (SSA and g) when AOD is that low, presents a minor role (please see the new Appendix A).

Page 8785 Line 20: at 400 nm?. Fix it.

++ Corrected, 440nm.

Page 8785 Line 25: Why do you use the fixed values of SSA (0.90) and g (0.75) for the cases with AOD<0.15 at 440 nm? I think you must explain it in the text due the representatives of the data in the region.

++ We included in the manuscript an Appendix about this choice. As you can see, these two values are not significant in the evaluations. As actual aerosols are in most cases mixed of different types, we think these values can be more representative, but this choice is not relevant in the evaluation of net fluxes. This choice was made attending to typical values of continental, maritime, and desert aerosols (see, e.g., Hess et al., 1998).

Page 8786 Line 3:

Which time interval is the "investigated period"? I think you must explain it, you mention in the abstract a period between 2004 and 2012, but there are different time intervals for all stations (Table 1).

++ We cancel 'during the investigated period', it is not relevant here. The time period in the abstract refers to the "mean database" for the Iberian Peninsula and it refers to 2004-2012, the individual datasets present the time periods indicated in Table 1.

Page 8786 Line 10 and Page 8787 Line 1: These sentences are the same, could you fix it?

++ Corrected. The first sentence was changed to: " For this purpose, cloud-free simulations are carried out by means of a radiative transfer code."

Page 8787 Line 13: You mention "In each interval, these properties are considered as wavelength independent." Which implications have this assumption to the calculations? How do you manage this consideration in the NIR interval when you have two values?

++ You can find a complete discussion of this topic in the new Appendix A. As you can see, the daily values present a relative difference between the spectral and fixed assumptions below 5% for most of the cases.

Section 4

Page 8788 Line 21, Page 8789 Line 6, 16, 22: I suggest to change the term "climatology" by other term like statistical or simply the annual behavior.

++ Following the reviewer's suggestion, the new title of Section 4 is "Analysis of aerosol properties over the Iberian Peninsula".

Page 8791 Line 8: You establish that "The temporal trend of aerosol load can be established over the last decade in the Iberian Peninsula". You have only one station with 10 years, El Arenosillo, the others stations have less or equal than 9 years. How could you establish the trend in one decade if you have not 10 years of data? There are stations with missing data in the time interval.

++ The trends are obtained per year, and 'per decade' is obtained *10. We decided to use per decade, because the numbers are easier to read. In order to clarify this issue, we added the following explanation to the manuscript: " The trends calculated in this study are obtained in the corresponding physical units per year. However, to unify notation with previous studies dealing with the radiative effect trends of clouds and aerosols (e.g., Mateos et al., 2013a), the results are multiplied by 10 and expressed in physical units per decade. In this way, the trends are also easier to read."

Furthermore, in Table 1 there was a typo. Also Palencia site has 10 years of aerosol data (2003-2012). Hence, one northern (Palencia) and one southern (El Arenosillo) sites can be representative for the Iberian Peninsula.

Figure 4: show the yearly mean values of AOD, apparently for the Granada station there is not AOD value for the 2009. Also for the station Cabo da Roca there are not values for the 2008 and 2009 years. These are not explained in the text. How can affect it to the trend calculation? There are not discussions about the causes of the trend in the Iberian Peninsula.

++ The Sen's method can be computed with gaps in the database. We justified this in the new paragraph about temporal trends*. The discussion of the possible causes is included in the Section 4.

* "With respect to the temporal trends calculated in this study, the Sen's method (Sen, 1968) is applied to evaluate the slope of a time series using the Mann-Kendall non parametric test to determine the significance of these rates. The Sen's method is not greatly affected by outliers and can be computed when there are gaps in the database (Collaud Coen et al., 2013). This is a common and adequate method in temporal trend evaluation (e.g., Sánchez-Lorenzo et al., 2013). The trends calculated in this study are obtained in the corresponding physical units per year. However, to unify notation with previous studies dealing with the radiative effect trends of clouds and aerosols (e.g., Mateos et al., 2013a), the results are multiplied by 10 and expressed in physical units per decade. In this way, the trends are also easier to read."

Section 5

Page 8791 Line 22: The title has the words inter annual and intra-annual evolution different to the word climatology used in the Section 4. Could you use an uniform terminology? Could you take care of the words daily and yearly or yearly mean value of AOD?

++ The new title of the Section 4 is: "Analysis of aerosol properties over the Iberian Peninsula", and we tried to simplify the notation about yearly values.

Page 8792 Lines 1-2: You mention ARENIR presents a more stable pattern, based in the results show in the figure 5. I don't see this more stable pattern, the pattern is similar in the three spectral bands with inter-annual changes.

++ This sentence is re-phrased. According to the reviewer's comment, the inter-annual changes are mentioned. " The patterns of ARE in the UV, VIS, and NIR ranges are similar with notable inter-annual changes. "

Page 8792 Lines 9-10: you mention the contribution of the slight reduction in the radiative effects of the atmospheric aerosol. But you do not discuss the reasons for this small reduction of ARE for the individual stations.

++ In the new version of the manuscript, the possible reasons behind the decrease in the AOD in the Iberian Peninsula and the consequent reduction of the aerosol radiative effects are mentioned. This discussion has to be performed at a "regional" scale, since the phenomena involved occur at the whole Iberian Peninsula.

Page 8795 Line 5: You write the phrase "AFENIR shows the weakest effect caused by aerosol absorption". Could you give some discussion about it?

++ This sentence is drawn from Table 3 (average values in different SSA categories), but looking at the SSA and alpha classification in the Figure 8, we decided to cancel this sentence, it is obvious that the SSA also plays a determinant role on AFEnir for each alpha interval.

Page 8795 Lines 6-21: You describe some reports AFE but you do not discuss the relation with the results in the work.

++ In the new table 4, you can find summarized the previous results. In this new version, it is easier to compare the results than in the previous version. We emphasized the different time periods, and therefore, the database used in each work. The larger discrepancies with respect to previous studies are due to the analysis of particular cases in those studies (our studies uses six long-term databases).

Referee 2

General comments

This paper by Mateos et al. presents data on aerosol optical properties and direct radiative effect (ARE) obtained at different Iberian sites in the long-term period 2001-2012. The main aim of the paper is to analyse the trend in aerosol content, properties and their radiative effect during this time interval in order to provide an aerosol climatology over the whole Iberian Peninsula. The ARE has been calculated separately in four spectral regions (UV, VIS, SW, and NIR) and its dependence on the absorption properties and size of particles has been investigated. The objective of the paper is appealing, however I have several comments about data analysis, discussions, and presentation which are reported in the following.

One of my main concerns regards the discussion of the results, which appears very poor and restricted to basic considerations. For instance, in Section 4 last paragraph, you present the results of Figure 4 without practically providing any comment. What is the cause for the trend? The reduction of emissions in Spain? The change in dust outbreaks occurrence? Is there a seasonality in this trend which may help to understand? Is there a connection with climatic indices (NAO, for instance) which may explain part of the interannual variability? Have you investigated that? Similar consideration are related to Section 5, where the discussion does not provide additional elements.

++ Following the reviewer's suggestion, we added new topics to the discussion. The possible causes of the reduction in the aerosol load are given, together with recent studies which support these findings. The relationship between climatic indices and aerosol load goes beyond of the scopes of this study, but we mentioned one particular year which has been proved as very interesting for the atmospheric science community.

My second main concern regards the fact that part of the ARE analysis and discussion does not provide neither new methods nor results. The obtained ARE values and the dependence on SSA, as also discussed by the authors, are in good agreement with several other studies performed in the Mediterranean basin. So basically the results of this paper confirm things that we already know. In my opinion, the most original and interesting part concerning ARE estimates is the discussion of the different spectral contributions (UV, SW, Vis, NIR), which unfortunately I have found only at the end of Section 6. I suggest the authors to consider reorganize the discussion around ARE estimates, especially in Section 6, in order to better highlight their findings.

++ In order to compare our results with the previous studies, new Table 4 summarizes the findings about AFE values. As the reviewer can see, the time periods of previous studies are shorter, being in most cases only referred to case studies. In our study:

- We highlight the AOD decrease in the 2000s in the Iberian Peninsula, which is relevant to understand the SW radiation increase (the brightening phenomenon) observed in this area (see, e.g., Mateos et al., 2013a). To our knowledge, this is the first study showing the AOD trends at several aerosol sites simultaneously in several sectors of the Iberian Peninsula.

- The method followed in this study with the aerosol properties simplifies the spectral behavior and the evaluation of ARE can be carried out with high accuracy.

- The ARE trends are also obtained (with a high significance level) for the "mean database" in the Iberian Peninsula 2004-2012. Again, this relevant result is necessary to analyze the brightening period.

- The used long-term databases have produced a detailed evaluation of ARE depending on aerosol properties.

- As the reviewer stated, the evaluation by intervals offers the possibility to perform a more complete discussion of aerosol radiative effects.

Third point, the analysis of uncertainties is not completely developed. For instance, can you provide error bars in Fig 4.? Also, it is not clear how the ARE uncertainty has been estimated. A certain number of assumptions have been performed to implement model calculations (Sect. 3), however the possible effect of these assumptions on the calculated ARE is not investigated (see for instance the specific comments below). To assess these uncertainties is however necessary to better constrain your results. Finally, for what concerns the presentation of data and results, I find that the paper is quite repetitive in some parts, Sections 4 and 5 in particular. Also, I have the impression that Figs. 4-5-6-7 have in part similar "messages", so probably they could be reorganized in order to merge them into 2-3 figures only.

++ We included the new Appendix A in the manuscript, where the reviewer can find the justification of the choices. The results shown in the Appendix support our methods.

We consider that all the figures are necessary because the information they highlight is different depending on the discussion. For instance, Figure 4 shows the yearly evolution of the AOD values at the six sites, before the average series is presented in Figure 6. Hence, we considered that these two figures must be shown in the article. Figure 6 is a summary of Figure 5 considering the average series, and the temporal trend rates are also presented. We decided to maintain all the figures as in the previous version, with the modifications suggested by the reviewer.

Specific comments

Introduction

I find that the scientific context and the main objectives of your study are not very well constrained. In particular, the second paragraph is quite confused; it seems for instance that you are interested only on dust, while also other aerosol types are investigated in the paper. I suggest you to revise this part.

++ According to the reviewer's suggestion, we changed this paragraph focusing on three aerosol types. "With regards to surface SW radiative effect (ARE_{sw}), Di Biagio et al. (2010) obtained the maximum radiative daily effects for different aerosol types in the Central Mediterranean in the period 2004-2007: -61 Wm⁻² (desert dust aerosols), -26 Wm⁻² (urban/industrial - biomass burning aerosols) and -43 Wm⁻² (mixed aerosols). All these negative figures point out a cooling of the Earth's surface. Aerosol radiative effects in the LW (ARE_{LW}) are expected to be smaller than in the SW and with positive sign (see, e.g., di Sarra et al., 2011; Antón et al., 2014). "

Section 2, pg. 8785, line 19: this 1% difference should be added to the SSA uncertainty

++ Not necessary, the relative difference between the two databases is 1%, and it is in the range of the uncertainty given by the inversion method of SSA. Therefore, the level 1.5-filtered data seems adequate to perform our study.

Section 2, last paragraph: how the SSA 0.90 and g 0.75 have been chosen? Can you add references for this? How can you state that this choice "provides a good characterization of the aerosol absorption"? Can you evaluate the uncertainty on your estimated ARE based on this assumption? Have you performed sensitivity tests to support your statement? Moreover, in line 27 I would avoid the expression "we think".

++ In the new Appendix A we analyzed this choice. The results indicate that the values of SSA and g under clean conditions (low turbidity) are not relevant.

Section 3, pg. 8786, lines 12-13: I do not understand the meaning of this sentence? It means that in the cited papers (Bilbao 2011 and Mateos 2013) the authors provide with comparison of modeled irradiances with measured data?

++ We changed this sentence: " The libRadtran model (Mayer and Kylling, 2005) has been shown to be a useful tool for obtaining solar radiation data, presenting high accuracy in both cloudy and overcast conditions (e.g., Mateos et al., 2013b, 2014; Román et al., 2014). "

Section 3, pg. 8787, lines 13-14: you assume wavelength independent optical properties in the different considered spectral intervals. Can you provide an estimate of the uncertainty induced by this assumption?

++ The new Appendix A includes this discussion.

Section 3, pg. 8788, lines 11-12: I would eliminate "daily" since the relation is general.

++ Canceled.

Section 3, pg. 8788, lines 11-17: there are several repetitions in this paragraph, please rewrite it in a more concise and clear form.

++ Following the reviewer's suggestion, the paragraph has been rephrased: " The aerosol forcing efficiency (AFE) is defined as the rate at which the radiative effect varies per unit of AOD (e.g., Di Biagio et al., 2009; and the references therein). The linear relationship between aerosol radiative effect and AOD is well known (see, e.g., Costa et al., 2004, 2006; Di Biagio et al., 2009). Hence, in this study, ARE is obtained as the slope of linear fits in the ARE vs AOD_{500nm} relationships. Therefore, AFE values are expressed in W m⁻² per AOD_{500nm}-unit (Wm⁻²τ⁻¹)."

Section 4 and Figure 4: can you specify the number of datapoints or measurement days for each year? Are they uniformly distributed throughout the different seasons for the different years? What about cloud cover? It is possible that some differences in the annual values reported in Fig 4 are related in part to specific episodes, such as for example an enhanced cloud cover during specific periods which has affected CIMEL measurements?

++ We added a comment about this topic to the manuscript. The aerosol measurements of CIMEL are only performed under cloud-free conditions, and this fact can produce that some events are only visible in some stations.

Section 4, pg. 8790, lines 5-15: how your classification is in agreement with the selection criteria by Toledano 2007 and Pace 2006?

++ We better explained this topic.

In line 14, what does it mean that "the value could be adjusted to the site"? How? Line 15, there is a repetition.

++ In studies dealing with previous studies, each station present different threshold for the classification using the alpha-AOD diagram.

"Note that the limit of AOD_{440nm} < 0.2 is arbitrary and this value could be adjusted according to the sites, which likely produce a different distribution different distribution in the pie diagrams. Actually, even close stations can present slight differences in the α -AOD classification (see, e.g., Obregón et al. 2012). "

Section 5, pg. 8792, lines 1-2: I do not agree when you say that in the NIR the ARE seems more stable; I have the impression that there are not significant differences between the different plots in Fig 5.

++ We changed this conclusion, we focused it on the inter-annual changes.

Section 5, page 8792, lines 26-27: I guess the larger contribution in the visible is due to the fact that the max of the solar spectrum is found around 700 nm.

++ As the ARE is obtained as a difference between two SW radiation values, the maximum of VIS range in each SW radiation could not be true in the ARE. This is the reason why we emphasized the maximum in the VIS range.

Figures 4 and 6: I suggest adding error bars in the plots.

++ We added to the plots the error bars for one station and one variable, respectively.