1 Dear Editor,

2 please find below the answers to the remarks of the reviewers. We want to thank to the reviewers for
3 they constructive comments.

4 Our replies are highlighted with red font and ** symbols.

- 5 The authors
- 6

7 <u>Referee #1</u>

8 General comments:

9 Mateos et al., 2014 presents aerosol radiative effect (ARE) obtained at six Iberian sites in different period for

10 each station covering from 2000 to 2012. Also, the aerosol forcing efficiency (AFE) is obtained. Both

11 magnitudes were calculated in the UV, VIS, NIR, and SW spectral regions. The paper addresses relevant

12 scientific questions within the scope of ACP. The principal objective of the paper is to analyze the behavior or

13 trend in aerosol optical properties and its radiative effect to produce a characterization of the aerosol over the

- 14 Iberian Peninsula.
- 15 The authors successfully achieved a manuscript with better quality than the previous version. They completed

16 the corrections suggested by the reviewer. I have only some suggestion of minor technical corrections,

17 outlined below. I propose to approve the paper for publication, after minor technical corrections.

- 18 Specific comments:
- 19 No sense of summation with de in equation 2
- 20 ** With the summation, we want to highlight that the sum of all the hourly values is divided by 24. We prefer
- 21 to maintain this notation, since the equation can create confusion.
- 22

Line 266-267 repeated phrase "different distribution"

- Line 266-267 repeated phrase "different of ** The repetition was cancelled.
- 25
- 26 Line 272 "...is in line..." ???
- 27 ** Corrected, "The classification used here is in line with...".
- 28
- 29 Line 961 and 968 Figure 8 and 9 axis X title "...group"
- 30 ** The titles were changed to "SSA".
- 31
- 32 In the section "references" there are some "et al.", could you complete the list of authors?
- 33 ** We've added all the co-authors.
- 34
- 35

36 <u>Referee #2</u>

37 **(our new comments are highlighted with ** and red font)

38

39 General comments

The revised manuscript by Mateos et al. has only in minor part improved compared to its first version. The authors have not satisfactorily addressed all my previous comments. It remains several open questions and doubts concerning the analysis and the results, which are detailed in the following.

- 43
- 44 At first, some point-to-point comments to author answers (author answers are indicated with ++).
 - 45

** Thank you for your comments. We would like to mention that this study focuses on the Iberian Peninsula 46 47 which is a region affected by pollution, desert dust, and clean continental aerosol types. Hence, the analysis of 48 the six longest aerosol series of the Iberian Peninsula provides the climatology necessary to understand the 49 main findings of the paper. To our knowledge, no studies of global Iberian Peninsula have been addressed until 50 now. Therefore, we think that the current structure is the most adequate. Section 4 presents the main 51 properties of the aerosols over the Iberian Peninsula. Section 5 presents the climatology and evolution of the 52 aerosol radiative effects (ARE). And finally, Section 6 analyzes the aerosol radiative forcing efficiency (AFE), 53 which needs Section 5 to be understood.

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56 REVIEWER OLD COMMENT: My second main concern regards the fact that part of the ARE analysis and 57 discussion does not provide neither new methods nor results. The obtained ARE values and the dependence 58 on SSA, as also discussed by the authors, are in good agreement with several other studies performed in the 59 Mediterranean basin. So basically the results of this paper confirm things that we already know. In my opinion, 60 the most original and interesting part concerning ARE estimates, is the discussion of the different spectral 61 contributions (UV, SW, Vis, NIR), which unfortunately I have found only at the end of Section 6. I suggest the 62 authors to consider reorganize the discussion around ARE estimates, especially in Section 6, in order to better 63 highlight their findings.

64

++ 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.

68

REVIEWER NEW COMMENT: I do not agree with this since many of the other studies refer to multiyear data (3
to 9 years), so covering a similar time period.

71

**As the reviewer can see in Table 4, the longest databases were used by Di Biagio et al. (2010), who covered
the period 2004-2007 for three different types of aerosols, and Valenzuela et al. (2012), who analyzed only
desert dust episodes between 2005 and 2010. Other articles covering more than 3 years only use a small

75 number of cloud-free days in their analyses, as it is indicated in the table (García et al., 2014; Esteve et al.,

- 2014). Therefore, our study provides a multiyear (with the longest aerosol databases of the Iberian Peninsula)
 and extended aerosol classification. To our knowledge, no studies with these longer data series at this number
 of stations is made in the Mediterranean basin and in other parts of Europe. The Iberian Peninsula is one of
 the really few areas with such number of close stations and long and good data series.
- 80 81
- ++ 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.
- 84
- REVIEWER NEW COMMENT: This is not so evident, since the accuracy of the ARE results depends on the
 accuracy of the used aerosol properties. As discussed in Appendix A your assumptions introduce an important
 level of uncertainty.
- 88
- 89 ** From the reviewer's comment, we think that the sensitivity analysis of the Appendix A focused on the clean 90 cases (AOD_{440nm} < 0.15) maybe was not clear enough. Those uncertainties were calculated analyzing the most extreme cases for the SSA between 0.8 and 1.0, hence the "real" uncertainty is smaller than this huge interval. 91 In fact, we have decided to change this discussion. The approach of using SSA = 0.9 and g = 0.75 is proved by a 92 93 comparison with four scenarios: 1) SSA = 0.80, g = 0.65; 2) SSA= 0.80, g = 0.80; 3) SSA = 1.0, g =0.65; and 4) SSA 94 =1.0, g = 0.80. These limits of SSA and g cover the large majority (even all) the values of the aerosol properties 95 over the Iberian Peninsula. As the reviewer can see in the new Table A.1, the uncertainty of the net fluxes is 96 small. We want to point out that the aerosol properties retrievals (SSA or g) under conditions with AOD_{440nm} < 97 0.15 present really large uncertainties and they are not reliable data. For instance, level 2.0 algorithms of 98 AERONET need an aerosol load > 0.4 to evaluate with guarantees these properties. Hence, our simple 99 assumption is proved as an adequate tool to consider the 70% of the aerosol conditions in sites such as 100 Palencia, Granada, or Évora. If these cases are omitted, no real characteristics are being analyzed.
- 101
- 102
- ++ The used long-term databases have produced a detailed evaluation of ARE depending on aerosolproperties.
- 105
- 106 REVIEWER NEW COMMENT: Not sure, see the previous comment.
- 107 ** This comment is solved with the previous replies.
- 108
- 109
- OLD COMMENT: 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 constraining your results.
- 115
- ++ We included the new Appendix A in the manuscript, where the reviewer can find the justification of thechoices. The results shown in the Appendix support our methods.

- 118
- 119 REVIEWER NEW COMMENT: The sensitivity study provided in Appendix A is correct and allows quantifying the
- 120 impact of the assumed optical properties on model calculations. Concerning this, I have 2 comments.
- 121 1. Line 498: add an appropriate reference
- 122 ** We added a reference.

124 2. The results of the sensitivity study indicate that there between 2 and 10% difference in the simulations of 125 the net spectral radiative fluxes as a function of the SSA and g values used in the calculations (deviations from the 0.90 and 0.75 values fixed for AOD<0.15). I consider these deviations absolutely NOT negligible, because 126 127 comparable to the absolute values of the forcing obtained by model calculations. Which is the ARE results 128 (value +/- uncertainty) if you take into account for this 2-10% error (the paper misses from a rigorous ARE 129 error analysis in Section 3, lines 197-214)? These uncertainties on the ARE retrieval at AOD<0.15 (which 130 represent a very large fraction of your data) largely impact the significance of the results shown in Figures 5 to 131 9). As a consequence, the results both in term of absolute values, seasonality, and temporal trends are largely 132 affected by that. A possible solution would be to distinguish data with AOD>0.15 and AOD<0.15 and to look at 133 what you obtain in this case.

134

135 ** This comment is solved with the previous replies (see the previous answer about Annex A calculations).

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REVIEWER OLD COMMENT: 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?

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++ 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.

- 147
- 148 REVIEWER NEW COMMENT: you did not completely answer to the raised questions which instead are critical149 to understand the differences observed in Figure 4.

150 ** We understand the reviewer query and we tried to solve this criticism in the new version. We think that a 151 table indicating the number of points used each year does not add significant information to the manuscript. 152 The six ground-based sites and the time period used in the study needs a large table. Instead, new Figure 4 is 153 plotted with a significant variation. The size of the symbol is selected attending to the number of points used 154 each year, i.e.: the larger the number of points, the larger the symbol for that year. In this way, a quick look is 155 only required to see the different data numbers. As regards the seasonal distribution, the measurements of 156 CIMEL (cloud-free conditions) are more often performed during summer months due to the climatological 157 conditions of the Iberian Peninsula. However, this higher likelihood of cloud-free skies during summer months 158 is not directly joined to the CIMEL data. There are months with technical problems, and therefore, with gaps in 159 the data series. Overall, most of the data (>30%) are obtained during summer, >20% in spring, 20% in autumn,

- and <20% in winter. But these approximate values are not extensible for all the years and sites. Unfortunately,
 non-continuous databases in the experimental physics are common and it is a usual problem. We added some
- 162 sentences to the text to clarify this topic:
- 163 "In addition, possible technical problems and meteorological conditions (CIMEL aerosol data are recorded
- 164 under cloud-free skies) cause a non-equally distribution through the year. Overall, summer is the season with
- 165 the largest contribution of data, followed by spring, autumn, and winter. "
- 166
- 167 The figure with the yearly data number distribution is the following:



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173 REVIEWER OLD COMMENT: Section 5, page 8792, lines 26-27: I guess the larger contribution in the visible is
 174 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.

179

180 REVIEWER NEW COMMENT: what I meant is that since the max of shortwave radiation is found in the VIS, the
 181 aerosol-radiation interaction at these wavelengths is generally more effective than in the other spectral
 182 ranges.

183

184 ** We agree with the reviewer, we have clarified this fact:

185 "The larger contribution of the visible spectral region with respect to the whole solar spectrum was also
186 noticed by Bush and Valero (2003), and this is expected since the maximum of shortwave radiation is found in
187 this interval.".

188 189	Other specific comments
190 191	Introduction : There are still some problems in the Introduction. At first I would eliminate Lines 52-54, since this sentence is not necessary and imprecise.
192	** The sentence was cancelled.
193	
194	I would suggest replacing Forster et al. (2007) with Boucher et al. (2013) (new IPCC report).
195	** Replaced.
196	
197 198	Also, I find that the second paragraph is still not clear and imprecise. For instance, in lines 67-68 you should specify that you refer only to dust particles for which the LW effect is relevant.
199	** We have indicated this annotation.
200	
201 202	Lines 72-73: not clear, and in any case the solar zenith angle plays a role not only in the UV region. ** We cancelled the part referring to the SZA.
202	

204 Section 4: lines 233-245 and 277-290, is there some effect due to the different altitude of the 6 stations? Why 205 in Figure 4 only the Barcelona data present error bars (the same for Figure 6)? Paragraph lines 308-318: it is

very difficult to thrust in the AOD temporal trend from the data in Figure 4. Some reasons are:

207 - There are not error bars in the plots;

** We added only error bars for Barcelona site for clarity. If the six error bars are presented, the figure results
 not so clear (see Figure below), but we would have no problem in the change of the figure. The altitude of the
 sites affects the aerosol vertical distribution of the lower tropospheric levels, and this topic is beyond our
 scopes.



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- We do not know how many measurements days correspond to each year data at each station; also, how they
are distributed along the year? You did not answer to this in the revised version of the paper.

** In the new version of the manuscript, the reviewer (and any reader) can understand with a quick look the
number of points used each year (see new Figure 4). We think that the detailed information required by the

- 219 reviewer it is not necessary in the follow up of the manuscript.
- 220
- By looking at your results and considering the results of statistical anlyses, I would conclude that the trend is
 NOT observed, except at one or two stations;
- ** We are sorry but we don't understand this reviewer's comment. In the article was only ensured (and
- proved) that the reduction in the AOD is only observed at site Barcelona with the necessary statistical
- requirements. Other two sites (Palencia and Évora) also present a decrease in the AOD (although with a
 smaller significance level). Overall, we think that the decrease of AOD can be stated for the Iberian Penir
- smaller significance level). Overall, we think that the decrease of AOD can be stated for the Iberian Peninsula
 in the 2000s. To solve these problems, when the temporal trends are reported, the *p values* of these trends
 are also given.
- 220

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- 231 Sections 5 and 6: In general, the accuracy of the obtained ARE is highly impacted by the uncertainties
- associated to model calculations for AOD<0.15 data (see one of the previous comments regarding AppendixA). This raises many questions on the obtained results and associated discussion.
- ** We do not agree with this comment. We have shown that the maximum uncertainty in the worst possible
 scenario for those cases with AOD < 0.15 is <5% for the ARE_{sw} (see Annex A). As we stated in the article, these
 cases represent the large majority of aerosol conditions over the Iberian Peninsula. Hence, as our study is
 aimed as an analysis of "real" aerosol conditions, these "clean" cases must be considered. In particular, we
 think the uncertainty of these cases is not too far that the "normal" uncertainty caused by different
 parameters often used in this kind of studies. The methods and control that the authors have of the aerosol
 CIMEL data is the best way to ensure their reliability. For instance, the staff of the Atmospheric Optics Groups
- CIMEL data is the best way to ensure their reliability. For instance, the staff of the Atmospheric Optics Groups
 of the University of Valladolid manage RIMA network to perform CIMEL calibrations (belonging to European
- 241 of the oniversity of valiabolic manage kink hetwork to perform clivit242 sites) in the framework of the AERONET network.
 - 243
 - 244
 - Line 326, please add references.
 - ** The references are mentioned after the sentence. Gkikas et al. (2013) and Pey et al. (2013) are introduced
 by "For instance,...".
 - 248
 - 249
 - Lines 332-333: it is not clear, I suggest rewriting the sentence.
 - 251 ** The new sentence is simpler: "Hence, both columnar and surface aerosols have pointed out a decrease in
 - 252 the aerosol load over the Iberian Peninsula. "
 - 253

- 254 References: There is an excessive number of self-citations, also with several works not related to the present
- study. I have noticed that in the revised version some others have been added (like for exemple the Mateos et
- al. 2014 JGR paper on cloud properties, which I find out of the main topic of the paper). In addition, I feel that
- 257 there are some sentences which are not necessary but that have been added with the intention of including
- some citations (for instance, lines 158-160, 194-196).
- 259 ** We understand the reviewer query, and we have reduced the number of self-citations.
- 260

In conclusion, I think that the paper still needs substantial revisions and that in the present form it does not
 deserve publication. One of my major concerns regards the accuracy of ARE estimates, which are highly
 impacted by the performed assumption on aerosol optical properties. The main conclusions are not supported
 by the data, in particular the temporal trends which are very uncertain and difficult to trust. I would encourage
 the authors to review their analysis and then to consider resubmission to ACP or to another journal.

** We think that this new version solves all the criticisms described by reviewer#2. The small uncertainty of
the clean cases (AOD < 0.15) is better shown in the new Annex A. The statistically significance of the result is

indicated by the *p value*, in order to clarify this issue. As the main topic dealt in this study, a global view of the
Iberian Peninsula is performed at the first time with the six longest surface based aerosol series (a total of 55)

270 identified at the institute with the six longest surface based aerosol series (a total of 55 271 years of aerosol data have been used). The evaluation of the aerosol radiative effects and its efficiency at four

different spectral ranges provides an extensive, complete, and interesting manuscript.