Atmos. Meas. Tech. Discuss., 7, C3603–C3614, 2014
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Atmospheric Measurement Techniques Discussions

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Interactive Comment

Interactive comment on "Relationships between columnar aerosol optical properties and surface particulate matter observations in north-central Spain from long-term records (2003–2011)" *by* Y. S. Bennouna et al.

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Received and published: 7 November 2014

General Comments:

* About LIDAR and CALIPSO data

As metioned by the referee the use of a lidar would be useful to explore the differences in vertical profile that may in turn explain differences in AOD and PM annual cycle. With this respect it should be noted here that there are no ground lidar data are available in this area. However, satellite data such as those of CALIPSO may provide more insight to this issue. The CALIPSO climatology dataset (Winker et al., 2013) is mentioned in the conclusion section in the view of possible future work. Besides, in our opinion we must investigate the behavior of air masses at three levels (500 m, 1500 m, 3000 m) in order to explain the differences between PMx and AOD cycles. The results of an analysis relying on airmasses have been added in additional material, and these results are used in the discussion on the climatological cycles.

* About the differences in the seasonal cycle of PMx and AOD

The authors acknowledge that the absence of bimodality cannot be explained only by sunphotometer sampling as compared to that of PMx, which is also demonstrated when taking coincident days as there is no change in the results obtained (mentioned in the text). As highlighted by the reviewer, it is not possible to say that the availability of more years of sunphotometer could reveal the presence of a bimodality in the AOD seasonal cycle. Indeed as mentioned by the referee, other major factors explaining the differences between the shapes of the annual seasonal cycle of PMx and AOD are vertical layering of the atmosphere and also possible occurence of major aerosol events under cloudy conditions. However, the later is in part related to sampling. It is obvious that sunphotometer is strongly influences by cloud cover, and that to some extent data may be lost because of high cloudiness but no correlation between aerosol events and cloud cover may be found. Consequently, some part of the text in the paper paper, including in particular the abstract, section 4.1 and conlusions have been removed and modified in that sense. Besides as mentioned above, the differences between PMx and AOD cycles are examined in relation with the results of an analysis relying on airmasses.

* About the dust signature in the seasonal cycle of PMx

The referee is right when he points out a lack of data in this present paper to demonstrate that the bimodality of PMx seasonal cycle reflects the signature of desert dust, and actually the authors should acknowledge that desert dust outbreaks might only partly explain the seasonality of the surface measurements (Querol et al., 2009; also reported in Cachorro et al., 2013). It is true that the authors focused in excess about the influence of desert dust to the point that the first sentence of referee 1 is textually "The paper presents analysis of EMEP PM and AERONET AOD datasets to discuss the occurrence of desert dust events in the north-central area of Spain ", and this is actually not the aim of the article. Therefore, some parts of the text of the article have been substancially modified in order to clarify this point.

However, the authors believe they cannot ignore completely a number of works focusing on Spain and the Mediterranean basin which have already explored the fact that the PMx and AOD monthly statistics present a desert dust signature in their seasonal cycle. Among the most recent works, we would like to recall in particular those of Querol et al. 2009, Pey et al. 2013 and Toledano et al., 2007. Hereafter we provide a number of quotes from these papers which are relevant in this aspect:

From Querol et al. 2009 :

Abstract : " As regards for the seasonal trends, these are largely driven by the occurrence of African dust events, resulting in a spring-early summer maximum over the EMB, and a clear summer maximum in the WMB, although in this later region the recirculations of aged air masses play an important role. Furthermore, a marked seasonal trend is still evident when subtracting the African dust load. This is characterised by a high summer maximum (driven by low precipitation, high insolation) and a winter minimum (intense synoptic winds). "

Page 4271 : " As regards the seasonality of African dust outbreaks in the EMB, Figs. 2 and 3 show clearly that the lowest frequency of African dust outbreaks occur from November to January; and that the maximum take place from late winter to spring, with a slightly lower frequency in summer. A similar pattern was observed for the WMB, but with summer episodes generally higher in frequency and intensity. The spring–

summer PM maxima are associated with the higher frequency of African dust outbreaks, lower precipitation, higher (local/regional) resuspension owing to the dryness of soils, increased formation of secondary aerosols caused by the maximum solar radiation, and recirculation of air masses that prevent air renovation (in the WMB). The sporadic second-order winter maximum in the WMB is attributed to intense pollution anticy-clonic episodes of anthropogenic PM and sporadic winter NAF episodes. "

Querol et al., 2009 considers that the maximum of spring in the WMB as sporadic but it is clear that it is a permanent characteristic of the climatology. Our works adds the climatology of the AOD and tries to explain the differences and analogies with that of PMx.

From Pey et al., 2013

" Mean African dust contributions to ambient PM10 concentrations across the Mediterranean Basin are varying according to the latitude. "

From Toledano et al., 2007 and Cachorro et al., 2013

Figure 1 (added in this answer to referee 2) shows the contribution of desert dust to the AOD annual cycle calculated for the AERONET sites of El Arenosillo in the south-western of Spain (left panel) and Palencia in north-central Spain (right panel), which are results taken from Toledano et al., 2007 and Cachorro et al., 2013 respectively. One can see that the absolute difference between the annual cycles considering all aerosol events and that considering all aerosol events excluding desert dust (hatched bars on the left panel and red bars on the right panel), present a bimodal shape. Due to its monthly distribution, the contribution of desert dust despite its small percentage, modulates the annual cycle of PM forming the minimum of April in the case of PM but not in the case of the AOD.

Figure 1 (added in this answer to referee 2)

caption: Figures from Toledano et al., 2007 and Cachorro et al., 2013 for desert dust

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contribution to the annual cycle of the AOD.

Besides it is important to remind here that the objective of this paper is not to give the evidence of the contribution from desert dust to the observed modulation, but only to establish the climatologies, make the comparison between them, and examine the relation between total column extinction with the AOD and ground surface level concentrations with PM measurements in this region of Spain. Therefore, most of information related with desert dust has been removed from the paper, and the climatologies of AOD and PMx are now explained by different causes, and in particular relying on the weights given by air masses origin (analysis of backtrajectories). These new results indeed suggest that the minimum of April in PM data is likely due to the absence of desert dust apportionment at the surface, consequently it appears that the shape of PM annual cycle is therefore modulated in relation with desert dust contribution. Concerning the link between bimodality and desert dust, it will be analyzed in details in two other papers that are currently in preparation. One of them is an extended version of the results on the dust inventory for Palencia/Peñausende published in a conference proceeding Cachorro et al. 2013, and the second paper is an analysis of the seasonality of the aerosol based on AERONET and PM data for the entire Iberian Peninsula. The results of this second study show that the bimodality is present from south to north, the remaining background seasonality (i.e. non desert dust days) nearly disapears in the southern part. As these papers have not been published yet and in the view of the comments of the referee, the authors therefore decided to avoid relying on those results and simply remove the statements in the text that are related to the latter which will be published afterwards.

* About the methodology applied for the detection desert dust events and used in the former version

For information, although the inventory of desert dust events in the region is based on a conference proceedings of Cachorro et al. (2013) written in spanish, it is important to note that the method applied in this study for the detection of desert dust events is the same as that used in a previous published paper Toledano et al., (2007b) focusing on the southwestern part of Spain with data from the AERONET site of El Arenosillo. The details on this method are already fully described in this peer-reviewed publication.

Specific Comments:

Page 5830 line1 :This change has been done.

Page 5838 lines 4-8 : The authors have added the graphs for the Angström exponent and PM2.5/PM10. However, this cannot be used to support this claim and therefore this statement has been modified in consequence. Actually, the contribution of desert dust must be studied in details and quantified in order to draw further conclusion. As mentioned in general comments, this is the object of another paper which is in preparation.

The authors have added the graphs for the Angström exponent and PM2.5/PM10 ratio (Figure 2 and 4), however it is important to notice that the annual cycle for Alpha (Angström exponent) or fine PM ratio (PM2.5/PM10) data directly obtained considering all days cannot be used here to demonstrate the influence of desert dust on the seasonal bimodality, as desert dust days reprensent only a small percentage of occurrence throughout the year, which contribute between 8 to 21 % to the AOD depending on the month. For this purpose, the annual cycles must be considered in the case of desert dust days and non desert dust days separately. Although the following figures are not added in this manuscript (for the reason mentioned before), they give the evidence that desert dust partly explains the observed bimodality. For this we show in Figure 2 (added in this answer to referee 2) the annual cycle of the AOD (440nm), PM10, Alpha(440-870nm) and fine PM ratio (PM2.5/PM10) considering only desert dust days on one hand (left panels a,c,e and g) and non desert dust days on the other hand (right panels b,d,f and h). As it can be seen for the days of non-desert there is an intrinsic bimodality in the climatology, the desert dust only reinforces this already existent behavior.

Figure 2 (added in this answer to referee 2)

caption: Annual cycle of the AOD (440nm), PM10, Alpha(440-870nm) and fine PM ratio (PM2.5/PM10) considering only desert dust days (a,c,e,g) and non desert dust days (b,d,f, h).

Page 5838 lines 26-29 : The nearby weather stations from the spanish national meteorological agency (AEMET) are located in the cities of Valladolid and Burgos. Although these stations do not provide cloud fraction observations, they provide climatological data for precipitation amounts and clear day count. In Figure 3 (added in this answer to referee 2), the climatological annual cycle of these two parameters is represented for all available meteorological stations of the " Castilla y León " region. As it can be seen precipitations show two minima, an absolute minima in summer (July-August) and a secondary minima in spring (March), separated by a maxima in May. This curve is obviously anticorrelated with that of clear day counts. As suggested by the referee, these type of data add insight into aerosol dynamics and possible meteorological co-variation with the aerosol in the region. Indeed, PM10 climatological annual cycle (Figure 2a in the paper) is anticorrelated with that of precipitations. However it is not the case for the AOD (Figure 2b in the paper) which does not present a secondary maximum in spring. This can be explained by the fact that precipitation regimes affect differently low and high layers of the atmosphere. Indeed, PM10 is related to low layers which are washed out by rain. The AOD being a column-integrated quantity, it depends on both low and high layers of the atmosphere. In the case of dust intrusions, dust layers above clouds cannot be washed out by rain. This element are now mentioned in the text in section 4.1.

Figure 3 (added in this answer to referee 2)

caption: Climatological annual cycle of precipitation amounts (top panel) and clear day counts (bottom panel) at different AEMET meteorological stations of " Castilla y León " for the period 1971-2000 (http://www.aemet.es).

Page 5839 line 13 : This was corrected in the text, and also in caption of Figure 4.

Page 5839, lines 17-20 : As regards the north-south gradient for PMx, the authors refer to the works of Querol et al., 2009 and Pey et al., 2013. Please see quotes provided in general comments. Moreover, the numbers (in percentage) for the contribution of desert dust to the seasonal cycle of the AOD have been published for the site of El Arenosillo in the southwestern area of Spain in the work of Toledano et al., 2007.

Page 5841 , lines 27-29 : The missing data points have been replaced by long-term averages because the method used to calculate the slope requires values for all months, and such a method was chosen because the data present a strong seasonal cycle. Leaving gaps and using another method which would have allowed to do so would not have changed importantly the results. For example, the estimation of the slopes using yearly means (calculated on all available daily data) give sensibly the same results. Therefore in this case the authors think it does not affect the results.

Page 5841 to Page 5846 : As this referee as well as referee 3 do not agree with the fact to refer to the conference proceedings of Cachorro et al., 2013 written in spanish, the authors decided to remove all the parts of the manuscript (text, Figures 6b and 7) relying on the results of this desert dust inventory. For this reason there are no answers to the following comments (see page-line list below) which concern parts that are not included in the new version.

Page 5841 , lines 23-26

Page 5842 , lines 19-23

Page 5842 , lines 24-25

Page 5843, lines 19-21

Page 5844, lines 3-4

Page 5844, lines 8-10

Page 5845, line 10-15

Page 5845, lines 23-24

Page 5845, line 25

Page 5846, line 3-6

Page 5847, lines 1-3 : The reference of Toledano et al., 2009 has been added.

Page 5848, lines 23-29 : The referee is indeed right, as shown in Figure 11d there is a good sensitivity to the parameter PM2.5-10. The contradictory statement mentioned by the reviewer has been removed (see answer to previous comment).

Page 5849, lines 15-18 : This is actually what the authors wanted to express, the sentence was rephrased for clarification to include the referee description in the text.

Page 5850, line 28 through Page 5850, line 2 : The text of the conclusions has been modified to take into account this important point, vertical profile being the major factor in the relationship between PM and AOD. This has been done also throughout the paper.

Figure 5 caption : Line 1 of the caption has been corrected as pointed out by the referee. Some text was also added for the identification of the anomalies and average value curves.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/7/C3603/2014/amtd-7-C3603-2014-supplement.pdf

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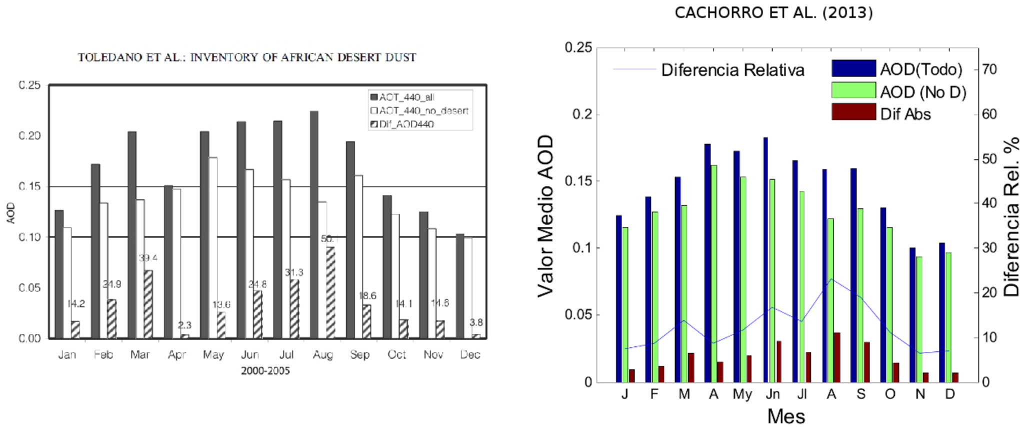

Fig. 1. Figures from Toledano et al., 2007 and Cachorro et al., 2013 for desert dust contribution to the annual cycle of the AOD.

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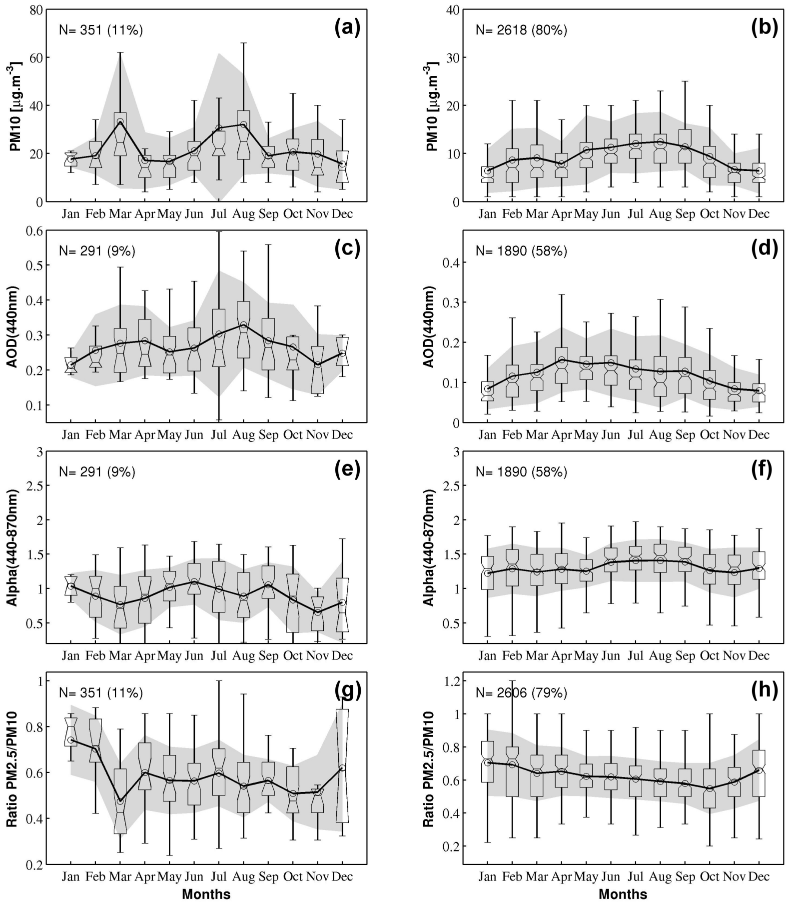

Fig. 2. Annual cycle of the AOD (440nm), PM10, Alpha(440-870nm) and fine PM ratio (PM2.5/PM10) considering only desert dust days (a,c,e,g) and non desert dust days (b,d,f, h).

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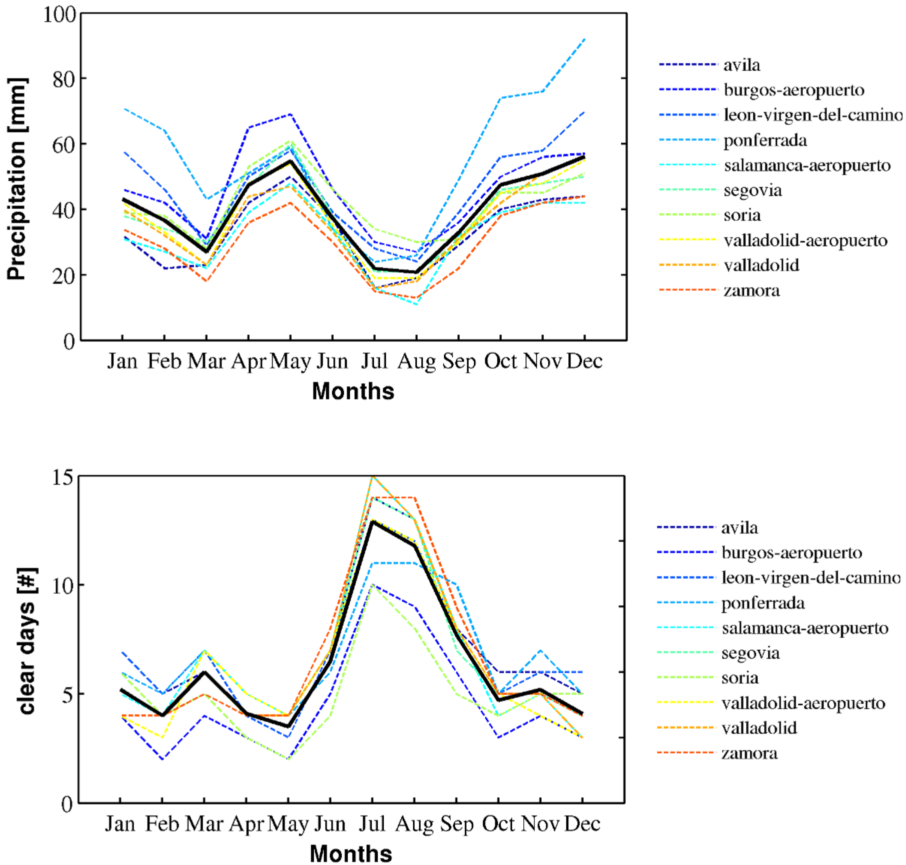

Fig. 3. Climatological annual cycle of precipitation amounts (top panel) and clear day counts (bottom panel) at different AEMET meteorological stations of Âń Castilla y León Âż for the period 1971-2000

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