

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.

Anonymous Referee #2

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Review for Atmospheric Measurements and Techniques Discussions

Title: Relationships between columnar aerosol optical properties and surface particulate matter observations in north-central Spain from long-term records (2003-2011)

Authors: Y.S. Bennouna, V. Cachorro, M.A. Burgos, C. Toledano, B. Torres, and A. de Frutos

General Comments: This paper presents interesting data of both Particulate Matter

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(PM) measured at ground level and Aerosol Optical Depth (AOD) which is representative of the total atmospheric column aerosol loading for sites located in North Central Spain. The paper presents some interesting results, however there is a lack of detailed discussion and analysis of the fact that the AOD is total column extinction versus PM measurements that are ground level mass concentrations in the current manuscript. There is no mention of ground-based lidar data or the satellite CALIPSO climatology (Winker et al., 2013) that would be useful to explore the differences in vertical profile that may explain differences in AOD and PM annual cycles. I cannot agree with the author's statements in both the Abstract and Conclusions that "In the case of the AOD this bimodality is likely to be masked because of poor sampling of sunphotometer data...", which implies that more years of sunphotometer data are needed to resolve the AOD seasonal cycle better. Other major factors that may result in seasonal cycle differences in AOD versus surface measured PM are the vertical layering of aerosol in long-range dust transport, and possibly a correlation between major dust events (and/or pollution/smoke) with cloud cover. Since PM measurements are made in all conditions including during cloudy conditions, the AOD data (which cannot measure when the sun is obscured by clouds) may be missing sampling of aerosol events associated with significant cloud cover. Additionally I find that there is a lack of data analysis to actually convince the reader that there is seasonality to the dust intrusions in this region. The authors suggest, that somehow the PM and AOD monthly statistics could show a seasonal dust signature. However both PM and AOD only indicate total aerosol concentration or mass not particle size, therefore seasonality in AOD and PM does not provide information on dust (coarse mode) monthly variation. I strongly suggest that the authors add a monthly plot time series of Angstrom Exponent and also PM2.5/PM10 statistics to be associated with Figure 2, in order to strengthen their now relatively weakly supported claims of seasonality in dust intrusions. Currently this paper primarily relies on the prior non-refereed work (conference proceedings) of Cachorro et al. (2013; published in Spanish) to define desert dust events (based on manual inspection of AOD and PM) in the data set, and additionally this data set is not

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sufficiently described in the current manuscript. I believe that time series of parameters sensitive to particle size (Angstrom Exponent and PM2.5/PM10) need to be shown and examined further in the current manuscript in order to strengthen the author's claims for dust causing the long-term trends and also average peaks in PM in March and August.

Some other aspects of the paper where I recommend revision or clarification are discussed below in the Specific Comments.

Specific Comments:

Page 5830 line 1: Please revise this sentence to include the words "total atmospheric column" before AOD and "ground-measured" before PMx.

Page 5838 lines 4-8: You should show both Angstrom Exponent and PM2.5/PM10 ratio time series to support this claim rather than just citing previous work.

Page 5838, lines 26-29: It would be informative to include a plot of AOD and also PM10 versus cloud fraction (cloud fraction observations from a nearby weather station). This might add insight into the aerosol dynamics and possible meteorological co-variation with aerosol in the region.

Page 5839, line 13: Fig. 1 should be Fig. 2 in the text.

Page 5839, lines 17-20: It would be useful to show a plot of the Angstrom Exponent and PM2.5/PM10 time series comparing the sites in the south versus the north of Spain in order to support your suggestion for a North-South gradient.

Page 5841, lines 23-26: These results suggest that there was no trend in dust contribution to PM, and therefore contradicts much of the rest of the paper. This is confusing.

Page 5841, lines 27-29: It is poor scientific practice to replace missing data points with long-term averages. It would be better to leave some gaps in the data than to insert average values.

Page 5842, lines 19-23: Much more detail should be given regarding the Cachorro

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et al. (2013) dust data set since this publication is not refereed and also not readily available to readers. Additionally it is written in the Spanish language, therefore many international investigators cannot read it. What Angstrom Exponent and PM2.5/PM10 ratios were used to identify dust in this conference paper?

Page 5842, lines 24-25: Please provide further explanation or discussion why this "desert dust data set" time series is NOT consistent with the time series of PM2.5-PM10 shown in Figure 6a which suggests no trend in dust near the surface (from PM differences data).

Page 5843, lines 19-21: Since AOD was moderate on 19-20 Mar 2005 you conclude that the AOD did not reflect the intensity of the suggested dust intrusion when PM10 was high. This seems a somewhat simplistic conclusion since you did not discuss the potential of vertical profile in causing the AOD versus PM discrepancy, and also since satellite images show much cloud cover on those days thus precluding AOD measurements. A more detailed meteorological analysis of this case is warranted.

Page 5843-5844, lines 29 – line 1 (next page): "Correcting" data points to make additional trend analysis is a poor scientific practice; how did you select the value of AOD=0.8 as a 'realistic' value?

Page 5844, lines 3&4: Change 'leeds' to 'leads'

Page 5844, lines 8-10: However this finding is significantly weakened by the fact that only an external data set from a non-peer reviewed publication allowed for this analysis.

Page 5845, lines 10-15: So, here you explain why you think the PM2.5-PM10 difference data are relatively useless, due to high uncertainty. However in Figure 11d the good relationship between Angstrom Exponent and PM2.5/PM10 suggests there is sufficient sensitivity to aerosol coarse mode fraction in both parameters. This contradiction needs to be clearly explained.

Page 5845, lines 23-24: You use PM2.5-PM10 on the x-axis of Figures 8 c and d,

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although in the previous paragraph you stated that this parameter was highly uncertain and therefore suggested it has little value. This is somewhat puzzling.

Page 5845, lines 25: Please provide further explanation and discussion to clarify why you concluded that a single aerosol type is likely to contribute to the coarse mode fraction. Why do you rule out sea salt and locally generated soil dust, and think that only desert dust exists in the coarse mode?

Page 5846, lines 3-6: Please state the main reason for the expected correlation between PM10 and AOD to be moderate. A likely primary reason is the variability of the aerosol vertical profile. Another is that small particles have a large influence on AOD but contribute much less mass than large particles to the PM10. Please expand your discussion here.

Page 5847, lines 1-3: Please give a reference here for typical clean continental region AOD and Alpha values.

Page 5848, lines 23-29: Figure 11d suggests that the PM2.5/PM10 ratio is sensitive to the presence of coarse mode aerosol relative contribution and therefore useful to detect desert dust occurrence. This seems to contradict previous statements that there is large uncertainty in a closely related parameter (PM difference; see page 5845). Please explain the apparent contradiction here.

Page 5849, lines 15-18: This 'delay' is probably better described as a time offset due to 24 hour sampling of PM at ground level versus less than 12 hour (seasonal change in day length) sampling of total column AOD from direct sun observation.

Page 5850, line 28 through Page 5850 line 2: Again, it is expected that this variability between surface PM and column AOD is expected largely due to variability in aerosol vertical profile. Vertical profile as a major factor in the relationship between PM and AOD and therefore needs to be mentioned here and also throughout the paper.

Figure 5 caption: Line 1 of the caption monthly 'means (right axis)' should be 'means

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(left axis). Also identification of the anomalies and average values curves is needed.

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