Aerosol properties over the western Mediterranean basin: temporal and spatial variability

By Lyamani et al.,

Answers to Dr. François Dulac

We would like to express our sincere gratitude to Dr. François Dulac for his helpful comments that will contribute to improve the quality of the manuscript. We have responded to each specific comment in detail and updated the manuscript according to his suggestions. Comments are in blue and our responses are in black, and the changes inserted in the manuscript are noted here in Red.

I am glad to see scientific valorization of data from the new AERONET station set up on the small remote island of Alborán during the project ChArMEx. Even though the measurement period is relatively short (~7 months), it reasonably covers the two Mediterranean summer and winter seasons. The analysis of this data set is complemented by (i) other coincident data from 3 AERONET stations in the westernmost Mediterranean region, (ii) a much longer time series from the relatively close station of Málaga, and (iii) shipborne data from a cruise across the Mediterranean and neighbouring marine areas of the Black Sea and northeastern Atlantic. I have some suggestions for a minor revision of the manuscript, which are following.

A point by point response is included below

Abstract:

-Line 4: I would name here the 3 additional AERONET stations (Málaga, Oujda, Palma de Mallorca).

Ok. We included the name of the 3 AERONET stations.

-Lines 12-19: I think the abstract would be better structured if the presently final discussion on Málaga data came just after Alborán results (before MAN results); this is because both Alborán and Málaga series appeal the same conclusion (which is not explicited in the present abstract but should be in my opinion since it appears as one of the main conclusions): the dominant role of long-range transport on the aerosol load at the regional scale in the westernmost Mediterranean region.

Following the reviewer 1 recommendations, we have removed from the abstract of the revised manuscript the discussion on Málaga data that focused on the evaluation of the impacts of the European ship emission regulations on the atmospheric columnar aerosol properties over this

station. In order to mention the role of regional pollution on aerosol load over westernmost Mediterranean sea in page 1 in lines 29-30 in the revised manuscript we added the following sentence "The fine particle load observed over Alborán was surprisingly similar to that obtained over the other three nearest AERONET stations, suggesting homogeneous spatial distribution of fine particle loads over the four studied sites in spite of the large differences in local sources".

Introduction:

-P.21525, lines 5-6: it is not correct that all satellite aerosol retrievals have low temporal resolution. MSG/SEVIRI AOD is available over the Mediterranean Sea with a 15 min temporal resolution ().

We agree with the reviewer that not all satellite aerosol retrievals have low temporal resolution. Thus, we have removed our statement about the satellite low temporal resolution from the new version of the manuscript.

-P.21525, line 7: cite a reference.

OK. We have included the following reference in the revised manuscript:

Estellés, V., Campanelli, M., Smyth, T.J., Utrillas, M. P., Martínez-Lozano, J. A. Evaluation of the new ESR network software for the retrieval of direct sun products from CIMEL CE318 and PREDE POM01 sun-sky radiometers. Atmos. Chem. Phys., 12, 11619–11630, 2012

-P.21525, line 23: "transported from European and North African urban"...

-P.21526, line 26: "westernmost part" rather than "western part".

Ok. We have corrected these sentences in the revised version.

-P.21527: in my opinion, one of the weakest points of the ms. is that there is no convincing argument that AERONET measurements are adapted to quantify ship traffic emission contribution to the column aerosol load. Unless you can argue the contrary, I would not emphasize this question.

Following all the reviewers' comments we have removed this part from the Introduction section of the revised manuscript. Also, we have removed section 3.5 that focused on the evaluation of the impacts of the European ship emission regulations on the atmospheric columnar aerosol properties.

-P.21528: name the three additional AERONET stations and give a minimum of information on the area covered by the MAN cruise considered. We have no information on the region covered in the Atlantic, which is a huge ocean. Plotting the cruise in a Fig. 1a would be useful.

In order to give information on the AERONET stations and MAN cruise covered area, in the revised manuscript, in page 4, lines 10-13, we have made the following change "Furthermore,

additional aerosol properties from three AERONET stations (Málaga, Oujda and Palma de Mallorca) surrounding Alborán Island and from a MAN cruise over the Mediterranean Sea, Black Sea and Atlantic Ocean (Figure 1) are analyzed....". In addition, according to the reviewer suggestions we included in the revised version the figure below where we show the location of Alborán Island, Málaga, Oujda and Palma de Mallorca and a MAN cruise track over the Mediterranean Sea, Black Sea and Atlantic Ocean (Figure 1) are analyzed...."

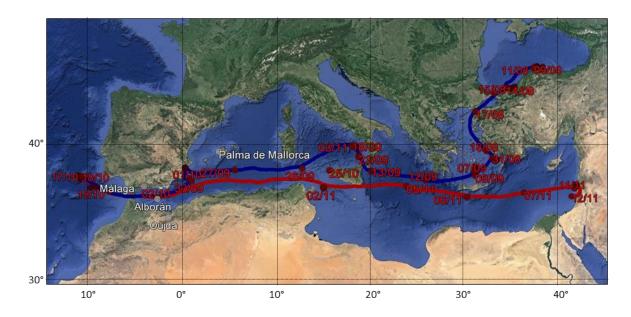


Figure 1. Map of Mediterranean basin showing the location of Alborán Island, Málaga, Oujda and Palma de Mallorca and a MAN cruise track over the Mediterranean Sea, Black Sea and Atlantic Ocean during 26 July-13 November 2011.

-Let me mention our paper from Mediterranean AERONET data which I believe should be referred to in the introduction and at several occasions in this manuscript: Mallet et al., Absorption properties of Mediterranean aerosols obtained from multi-year ground-based remote sensing observations, Atmos. Chem. Phys., 2013.

Ok. We have included the aforementioned reference in the revised manuscript.

Instrumentation and study sites:

-I suggest using sub-sections and restructure this section: 2.1 AERONET measurements including p.21529 lines 10-28 and p.21530 sentence on line 12-13; 2.2 AERONET stations; 2.3 Maritime Aerosol Network measurements; 2.4 Airmass trajectories.

We have restructured this section as recommended by the reviewer.

-p.21529, line 1: any ref. on the ship traffic and associated emissions?

Information on ships traffic can be found in (<u>www.marinetraffic.com</u>). Following the reviewer recommendation we included this information in the revised manuscript. On the other hand, unfortunately, we don't have information on the ship associated emissions.

-A table is missing to provide details on the AERONET data sets, including exact period, number of days (for each month) for each stations.

Following the reviewer suggestions, we have included the exact period for Alborán station in the revised manuscript in page 5 in lines 11-13 "This study focuses on the AERONET sun photometer measurements acquired at the Alborán Island (35.90^o N, 3.03^o W, 15 m a.s.l), in the western Mediterranean Sea, from 1 July 2011 to 23 January 2012." In addition, in page 5 in lines 31-32 we have included the exact period for the other stations "...we used AERONET data obtained from 1 July 2011 to 23 January 2012 over three AERONET stations surrounding Alborán Island; Oujda, Málaga and Palma de Mallorca."

According to the reviewer 1 we added the comparison of daily mean values of δ_a (500 nm) obtained at Alboran and Oujda (Fig. 6b) and those obtained at Alboran and Palma de Mallorca (Fig. 6c). We think that the number of measurement days for each station and each month can be easily inferred from these plots in figure 6.

-More details would be welcome on the MAN cruise. I suggest to reproduce the ship track with the different months of the period.

As we commented in the old version of the manuscript clear and more detailed information about the Nautilius_11 cruise track can be found in (<u>http://aeronet.gsfc.nasa.gov/new_web/cruises_new/Nautilus_11.html</u>). In any case, we have also included Figure1 in the revised version, showing the MAN cruise track over the Mediterranean Sea, Black Sea and Atlantic Ocean (see our previous response).

-Trajectories: they were probably computed only for days with AERONET measurements? Are they performed also for MAN observations? Did you check for forest fires along trajectories and did the photometers sample such events in their time series.

Backward trajectories were calculated only for days with AERONET measurements and also for days with MAN observations. To clarify this point in the revised manuscript in page 6 lines 27-30 we made the following change "5-day backward trajectories ending at 12:00 UTC at these sites for 500, 1500, 2500, 3500, 4500 and 5000 m above ground level were calculated using the HYSPLIT model for days with AERONET measurements (Draxler and Rolph, 2003). In addition, backward trajectories ending at the different points of MAN cruise for 500, 1500, 2500, 3500, 4500 and 5000 m above ground level were also performed for days with MAN observations."

This study mainly focuses on the AERONET sun photometer measurements acquired at the Alborán Island. In this sense, we have checked Alborán data in more detail (using information on forest fires provided by MODIS and air mass back trajectories analysis) in order to detect a

possible influence of biomass burning aerosol from forest fires. However, we didn't find any special biomass burning event during the analyzed period.

-Some information on precipitation occurrences at the sites could be useful.

Unfortunately, we don't have information on precipitation occurrences at all these sites.

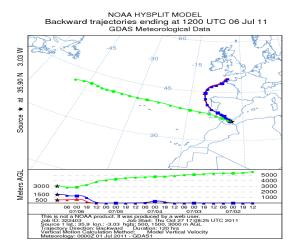
Results and discussion:

- δ C and δ F could also be discussed in 3.1.

Values of δC and δF are already discussed throughout section 3.1 at different part (see for example Page 7 lines 19-26 and page 10 lines 1-8. We think that additional information would be redundant.

-Bottom of P.21532: can you exclude that air masses from the Atlantic with low δa and low α contain some dust after long-range transport from NW Africa?

According to back trajectory analysis, air masses from the Atlantic with low δa and low α passed over Iberian Peninsula before reaching Alborán Island (see for example the figure below). However, during the analyzed period, these Atlantic air masses didn't travel over North Africa before reaching Alborán Island. Thus, we think that these Atlantic air masses may contain anthropogenic particles from Iberian Peninsula but can't contain desert dust from North Africa.



-P.21533: line 24: the MODIS image is not very convincing; it would be better to show AOD product, especially from MSG/SEVIRI; see for instance the ICARE ChArMEx multibrowse tool at <u>http://www.icare.univ-</u>

<u>lille1.fr/browse/?seviri_aer_oc_l2_tau=true&caliop_l1_exp=true&north=50&west=-</u> <u>10&east=40&south=25&size=large&date=2011_08_26&rebuild=false&pointer=zoom</u>.

OK. We included the AOD product of MSG/SEVIRI in the revised manuscript.

-P.21533, bottom: which fraction of days would be misclassified?

Here, our intention is to highlight that the use of $\delta a(500 \text{ nm})$ and $\alpha(440-870)$ thresholds for discriminating aerosol types may fail in some cases and that additional information will help obtain better identification of aerosol types. The statistical analysis of misclassified days is out of the scope of this work.

-P.21534, lines 10-15: can you really exclude dust particles in the fine mode?

Here we don't try to exclude the desert dust contribution to the fine mode. At this stage, we only try to highlight that the contribution of fine mode particles (either dust or anthropogenic or both) was more relevant during dust events associated with air masses transport at low levels (at 500m or 1500m level) from northern Italy/Mediterranean Sea than during desert dust events not associated with air masses coming from northern Italy or Mediterranean Sea. In order to avoid confusion in page 9 in line 33 we made the following change "These results highlight a considerable contribution of fine mode particles (either dust or anthropogenic or both) to the aerosol population (FMF ranged from 20% to 52%) during these dust events"

-P.21534, line 17: I would add "which indicates that the contribution of mineral dust to the fine mode fraction of AOD is probably significant".

As we commented in the old version of the manuscript, back trajectory analysis for days with dust intrusions with highest fine aerosol load revealed that the air masses reaching the study area at low levels (at 500m or 1500m level) come from northern Italy and the Mediterranean Sea. However, during desert dust events with lowest fine aerosol loads, none of the air masses affecting the study area come from northern Italy or Mediterranean Sea. These last cases include also desert dust events associated with air masses transport at all altitude levels (from 500m to 5000m) from North Africa. Thus, we think that these results point to the relevant contribution of anthropogenic particles to the fine mode fraction of AOD during dust events associated with large loads of fine aerosol particles and not to mineral dust. In order to clarify this point in page 10, lines 6-8, we added the following sentence "which points out significant contribution of anthropogenic particles to the fine mode fraction of δ_a (500 nm) during desert dust events associated with large loads of fine aerosol particles."

-P.21535, line 1: such formulation suggests that Italy should be the source, but central or eastern Europe can well be concerned.

Following the reviewer suggestions in page10, line 24, we made the following change "Thus, the desert dust transport appears to be a main cause of high aerosol loads while transport from central European urban areas is associated with occasional large aerosol loads over Alboran Island."

P.21537, line 26: it might well also be due to the fact that large scale pollution is dominating the region, independently of the contribution from ship traffic.

Following the reviewer suggestions in page 14, lines 3-5, we added the following sentence "These results suggest homogeneous spatial distribution of fine particle loads over the four studied sites in spite of the large differences in local sources."

P.21538, line 20: I think that Moulin et al. (JGR, 1998) first described this gradient.

OK. We have included this reference in the revised manuscript.

-Top of p.21542: rather check δC for dust; you could check for a diurnal cycle in the data in relation to the hypothesized breeze effects.

-P.21542, line 13-14: what about $\delta F?$

-Top of P.21543: you should check the ship traffic evolution since an increase in traffic might compensate lower emissions per ship.

-P.21543, line 9: not clear to me which type of data could effectively be used to apportion the European Directive effects. Please clarify your point.

According to reviewer1 suggestions we have removed section 3.5 from the revised manuscript.

Conclusion:

-P.21544, line 5: Recall what characterizes "background maritime conditions".

Ok. In page 17, lines 24-25, we made the following change in the revised manuscript "Background maritime conditions over Alborán characterized by low aerosol load and Ångström exponent (δ_a (500 nm)<0.15 and α (440-870)<1) were observed on about 40% of the measurement days."

Tables:

-Table 2: given the type of variability, geometric means would seem more adapted than arithmetic means to average the distribution of values.

The majority of studies report arithmetic mean values of aerosol properties. Thus, in order to compare our results with those reported by other authors (as we do in this study) we think that arithmetic mean values are more appropriate.

Figures:

-Fig. 1 could be complemented with a Fig. 1b plotting the MAN cruise track with the different months along the track.

Please, see our previous response.

-Fig. 2 is hardly readable, colour is requested to distinguish the 3 wavelengths; you should stretch axes at the maximum, possibly making 2 different large (full page) figure; I believe that a plot or a

2-D histogram (as in Paronis et al., Aerosol optical thickness monitoring in the Mediterranean, J. Aerosol Sci., 29, 1998) showing 2 as a function of 2A, would be very useful for data interpretation.

OK. In order to make Fig. 2 easy for data interpretation we made the change suggested by the reviewer and we only represented the AOD at 500 nm and 1020 nm (see figures below).

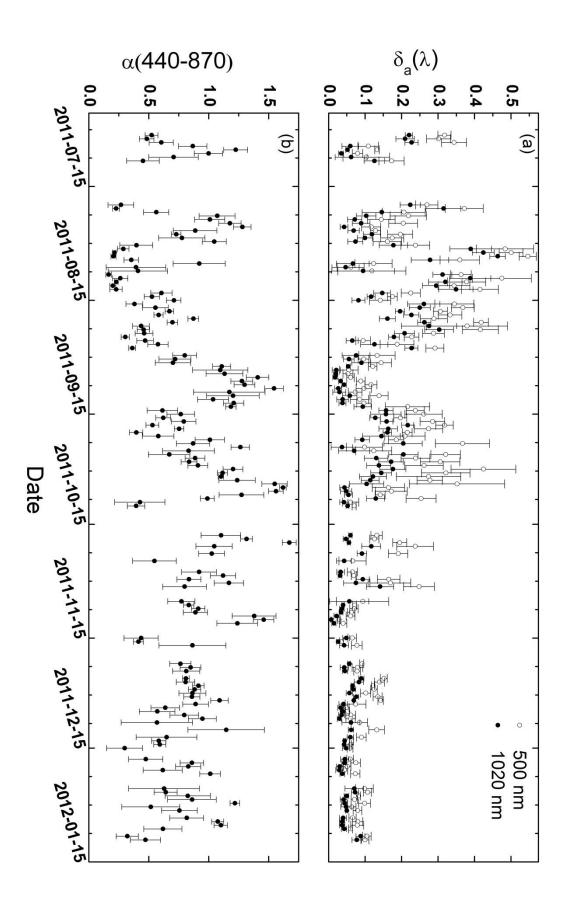
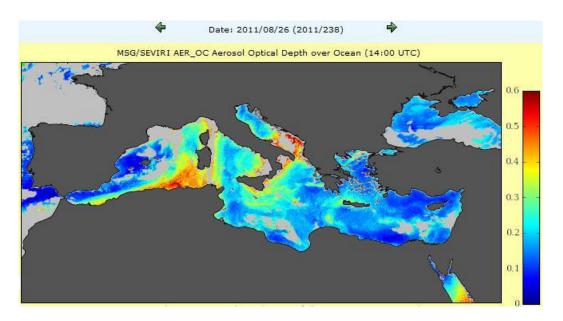


Fig. 3c: this MSG-derived AOD at 14 UTC) gives a much better evidence of the dust plume in the Alborán Sea (and not in Málaga) (see ICARE web site mentioned above; proper ref. for this product is Thieleux, F. et al., Remote sensing of aerosols over the oceans using MSG/SEVIRI imagery, Annal. Geophys., 23, 1-8, 2005):



OK. We included this MSG-derived AOD in the revised manuscript.

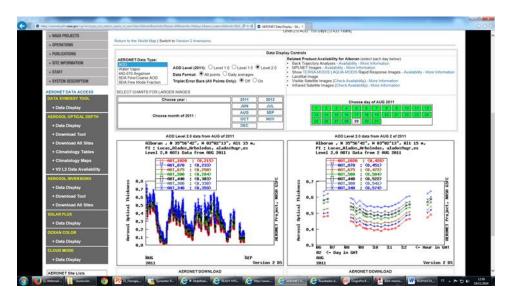
Fig. 5a: it seems that δa is not equal to $\delta F + \delta C$ in this plot.

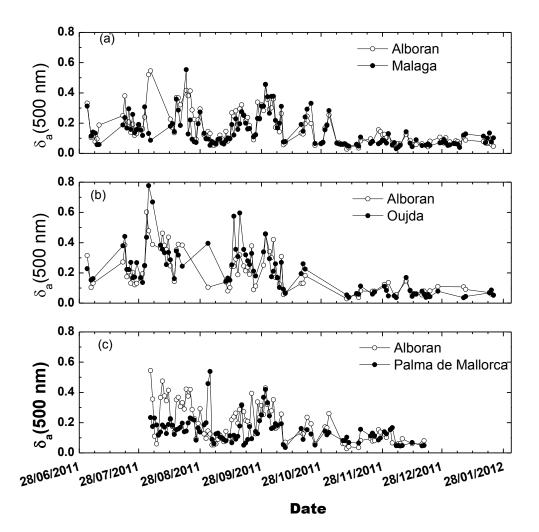
We have checked these data and we found that δa is equal to $\delta F{+}\delta C.$

Figure 6: I checked data on the AERONET web site and I did not find that level-2 data which are supposed to be used are corresponding to the plot shown in Fig. 6; I had the feeling that level 1.5 data might have been used but for Alborán, there is no data set that corresponds (in particular the early August intense peak of 2 days): please check your data set; intermediate ticks on the time axis would be helpful; it might be useful to add (as a Fig. 6b for instance) a correlation plot between the two series.

As we commented in the old version of the manuscript the AERONET data used in this work are level2 data. As can be seen in the figure below, Level2 AOD obtained in Alborán in early August was very high as in figure 6. However, it is important to note that the daily mean data presented in Fig. 6a were calculated only from time coincident measurements over Alborán and Málaga. On the other hand, following the reviewer suggestions we included the intermediate ticks on the time axis of this Figure. Also, following the suggestions of the reviewer 2 we added 2 figures (Fig. 6b and Fig.6c; see figures below) in the revised version of the manuscript where we presented the temporal evolutions of daily mean values of δ_a (500 nm) from July 2011 to January 2012 obtained over (b) Alborán Island and Oujda and (c) Alborán Island and Palma de Mallorca.

In addition, we provided information about the correlation plots between each two series in the new version of the manuscript.





Editorial comments:

- -P.21525, lines 19 and 24: lower case for "oceans and seas".
- -P.21525, lines 25: "forest fires" (plural).
- -P.21526, line 26: "westernmost part" rather than "western part".
- -P.21527, line 24: "there is no study".
- -P.21528, line 8: "a MAN cruise".
- -P.21528, line 10: "implementation".

-P.21528, line 24: "50 km" (with a space).

-P.21528, line 27: I suggest "There is no significant local anthropogenic emission source at Alborán".

-P.21529, line 11: specify "automated sun photometer".

-P.21530, line 19: specify "hand held sun photometer".

-P.21530, line 28-29: figures seem to indicate that GDAS meteorological data, not CDC1, are used; lower case for "meteorological data".

-P.21532, line 29: specify "during the wet season from November to July".

-P.21534, line 24: "there is no[...] activity".

-P.21535, line 17: "increase".

-P.21535, line 26: "there was no[...] intrusion".

-P.21536, line 6: "On the other hand".

-P.21536, line 23: specify "in comparison with the Alborán station".

-P.21537, line 24: "it is expected".

-P.21542, line 12: "the study site".

-P.21543, line 22: "than reported".

-P.21545, line 4: "other marine regions".

-P.21545, line 10: what about P??

-Table 4: last line in table should probably read "Number of coincident measurement days".

-Figure 4a and b are hardly readable: expand axes at the maximum, use colours for the different wavelengths; comment on the arrows in the legend.

-Legend of Fig. 5b: "Monthly relative frequency".

Thank you. We corrected all these typing errors in the revised manuscript.