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

By Lyamani et al.,

Answers to reviewer 1

We would like to express our sincere gratitude to the anonymous referee for his/her helpful comments that will help to improve the quality of the manuscript. A point by point response is included below. Comments are in blue and our responses are in black, and the changes inserted in the manuscript are noted here in Red.

The paper analyzes a data-set of radiometric data coming from the Alboran Island, located in Western Mediterranean Sea, far about 150 km from Malaga city and 50 km from North-Africa coast. Authors analyzed the period July 2011- January 2012. This can be a good observation point to study aerosol mixing effect of background aerosol with dust particles from Sahara, or with polluted emissions from ships crossing the Mediterranean Sea. To determine spatial and temporal variation of aerosol properties, authors complete their study comparing data from Alboran with coincident data from Malaga (Spain), Ojuda (Morocco) and Palma de Mallorca Island (Spain). Moreover they compare Alboran data with ship sun photometer measurements obtained during a cruise from the Black Sea to the Mediterranean Sea in the period July-November 2011, in the framework of the Maritime Aerosol Network (MAN). Finally, they try to derive information about the possible effects of the EU Directive on ship emissions from long-term radiometric data obtained in Malaga. This paper can be published on ACP under major revisions. In the following the critical points to be carefully revised.

A great part of the Introduction considers the ships as sources of air pollution, due to their SO2 emissions by which sulfate aerosol derive. However the paper, dealing with radiometric measurements only, do not consider SO2, neither sulfates data. For this reason, this part of the Introduction should be omitted or rewritten according to the treatment of this part in the corresponding paragraph.

Following the reviewer's recommendation we removed this part from the Introduction section. We also removed section 3.5 that focused on the evaluation of the impacts of the European ship emission regulations on the atmospheric columnar aerosol properties.

Page 6: authors describe the site characteristics and the possible sources affecting the atmosphere in Alboran. According to the regional circulation and in absence of local sources, they expect Alboran island to be affected by anthropogenic pollutants from Europe and desert dust from North Africa. They should consider also the Mediterranean Sea as an aerosol source.

OK. In the revised version, in page 5, lines 19-23 we made the following change "”Due to its location, Alborán Island is expected to be affected, depending on regional circulation, by anthropogenic pollutants originated in urban and industrial European areas, anthropogenic particles emitted from the ship traffic, desert dust transported from North African arid regions and maritime aerosols from the Mediterranean Sea”.

Page 9: Differences among mean values parameters obtained with present study and those reported in literature could depend, also, on different air masses circulation and different periods and duration of the measurements campaigns.

OK. In the revised version, in page 8, lines 14-16 we made the following change " The differences between aerosol properties observed over Alborán, Lampedusa and Crete Islands could be explained in terms of

differences in the period and duration of the measurements, in air mass circulation and in the methodologies employed."

Page 9: 40% is not a poor percentage of background marine conditions. Please, delete "only".

Ok.

Page 9: Looking at the work of Smirnov et al., 2002 (table 3), it can be seen that pure maritime conditions correspond to $\tau(500)<0.1$ and alpha< 0.8. The values considered by the authors of present work ($\tau(500)<0.15$ and alpha< 1) correspond to mixed maritime conditions. In order to verify the goodness of the Smirnov's criterion for pure maritime situations, they should examine measurements fitting the conditions $\tau(500)<0.1$ and alpha< 0.8.

In their 2002 paper Smirnov et al. (2002) only discussed measurements of aerosol optical depth in maritime and coastal areas; however, they did not present the analysis of pure maritime aerosol. Based on this work, Smirnov et al., (2003) presented an aerosol model for "pure maritime" aerosol considering the thresholds we use in this study, $\delta_a(500)<0.15$ and $\alpha(440\text{–}870)<1$. We apologize because there was an error in the reference given in the previous version of the manuscript. The correct reference is: "Smirnov, A., B. N. Holben, O. Dubovik, R. Frouin, T. F. Eck, and I. Slutsker (2003), Maritime component in aerosol optical models derived from Aerosol Robotic Network data, J. Geophys. Res., 108(D1), 4033, doi:10.1029/2002JD002701."

Thus, in the revised version, in page 8, lines 19-20, we corrected by:"According to the Smirnov et al. (2003) criterion, pure maritime situations can be generally found when $\delta_a(500\ \text{nm})<0.15$ and $\alpha(440\text{–}870)$ is less than 1. Considering this criterion, pure maritime situations were observed over Alborán Island on 40% of the analyzed days."

In any case, similar threshold values ($\delta_a(500\ \text{nm})\leq 0.15$ and $\alpha(440\text{–}870)\leq 0.6$) were used by Toledano et al. (2007) and less restrictive ones ($\delta_a(500)<0.2$ and $0.2<\alpha(440\text{–}870)<1$) were used by Sayer et al. (2012 a,b) for identifying pure marine aerosol. In this sense, in the revised version, in page 9 lines 3-7 we made the following change "Threshold values for $\delta_a(500\ \text{nm})$ and $\alpha(440\text{–}870)$ have been widely used in remote sensing to identify marine aerosol type. For example, Smirnov et al. (2003) used $\delta_a(500\ \text{nm})\leq 0.15$ and $\alpha(440\text{–}870)\leq 1$ and Sayer et al. (2012a,b) proposed $\delta_a(500\ \text{nm})\leq 0.2$ and $0.2\leq\alpha(440\text{–}870)\leq 1$ while Toledano et al. (2007) used $\delta_a(500\ \text{nm})\leq 0.15$ and $\alpha(440\text{–}870)\leq 0.6$ for identifying pure maritime situations.

The references below are included in the new manuscript version

Sayer, A. M., Smirnov, A., Hsu, N. C., and Holben, B. N.: A pure marine aerosol model, for use in remote sensing applications, J. Geophys. Res., 117, D05213, doi:10.1029/2011JD016689,2012b.

Sayer, A. M., A. Smirnov, N. C., Hsu, L. A. Munchak, and B. N. Holben.: Estimating marine aerosol particle volume and number from Maritime Aerosol Network data. Atmos. Chem. Phys., 12, 8889–8909, 2012.

The day under examination (26 August 2011) does not always fulfill this last condition.

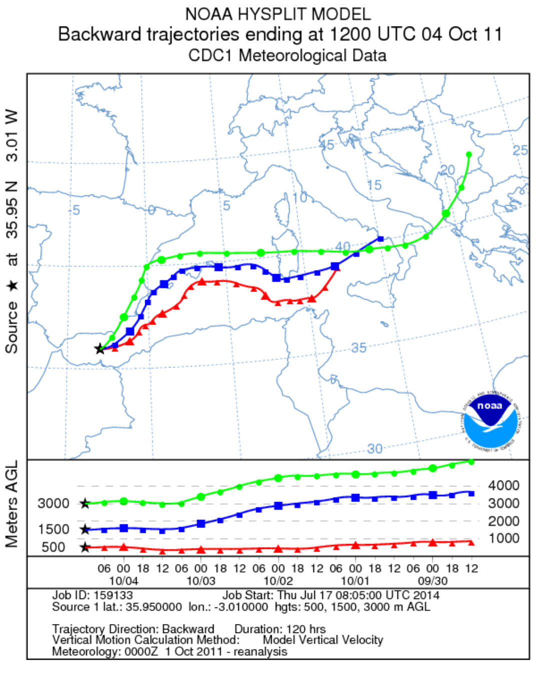
As we commented before we used the criterion ($\delta_a(500)<0.15$ and $\alpha(440\text{–}870)<1$) proposed by Smirnov et al. (2003) for identifying pure maritime aerosol. In this sense, the Smirnov et al. (2003) criterion is always fulfilled in 26 August 2011 as can be seen in Fig 3. In any case, the more restrictive criterion ($\delta_a(500\ \text{nm})<0.1$ and $\alpha(440\text{–}870)<0.8$) suggested by the reviewer's for identifying pure marine aerosol is also fulfilled for most of the measurements in this day. Particularly, this restrictive criterion is also fulfilled during satellite overpass over Alboran, when the satellite image clearly reveals the presence of dust over Alborán.

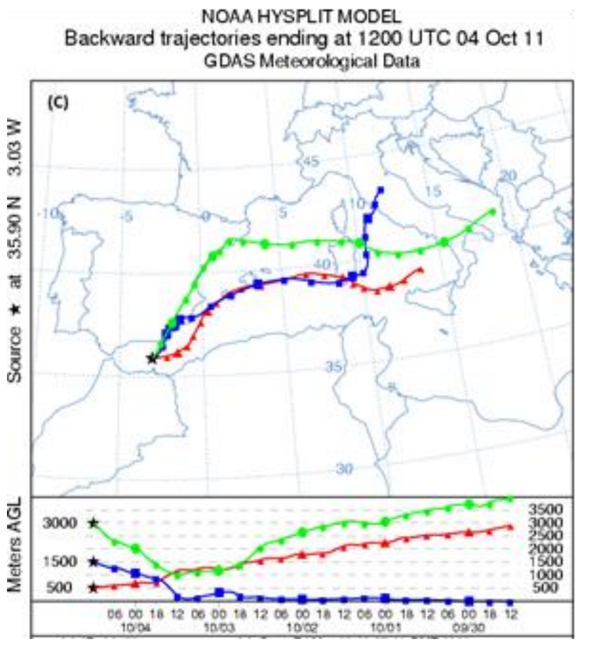
In any case," background conditions" should correspond, by definition, to low aerosol loading, that is very low AOD values. Moreover, due to the location of the measurements site, it would be expected to find marine aerosol as one of the major components of background conditions. It is interesting to note in figure 2a-b how conditions of poor aerosol loading in Alboran correspond to 1< alpha < 1.5. This could suggest that background conditions are actually characterized by marine particles, both in their fine and coarse components. Measurements of single scattering albedo, if available, could represent a useful tool to identify and classify optical particles properties.

We agree with the reviewer's that "background conditions" refer to low aerosol loading and that background conditions (δ_a(500 nm)<0.15) at Alborán may include polluted maritime in addition to pure maritime situations, as indicated by Ångström exponent values in the range 0.1-1.6. However, in this section of the manuscript our objective was to examine "pure" maritime situations (δ_a(500 nm)<0.15 and α(440–870)<1). We don't refer to background conditions which may include in addition to marine aerosol other particles from continental origin. In order to avoid confusion we changed "background conditions" by "pure maritime situations".

On the other hand, we also agree with the reviewer's that single scattering albedo could help to improve our classification of optical particles properties. However, single scattering albedo for low aerosol optical depths (AOD<0.2) presents high uncertainty (Duvobik et al., 2002). In any case, in our study only two instantaneous single scattering albedo values of level 2 were obtained during the analyzed period and thus we do not include analysis of single scattering albedo in this work.

On page 7 authors say they use HYSPLIT model with CDC1 Meteorological data, but figures 3b and 4c have been calculated with GDAS model. Authors conclude that measurements obtained from 30 September to 4 October are affected by polluted air-masses coming from North-Italy and report in figure 4c, as an example, HYSPLIT back-trajectories at 12:00 on 4 October 2011 with GDAS Meteorological Data. Looking at the figure this peculiarity do not appear, rather the trajectory ending at 1500 m suggests a strong presence of marine particles. Also the back-trajectories derived with the CDC1 data-set and reported in the figure below highlights a strong marine contribution to the aerosol load. For this reason authors should re-discuss this part of their comments.





In this study, we have used HYSPLIT model with GDAS Meteorological data. In the revised version, in page 6, line 31, we corrected this mistake and thus the new wording reads as follows:"The model version employed uses GDAS Meteorological data".

It is generally established that non-polluted maritime situations are typically characterized by low columnar aerosol loading (AOD<0.15) and by volume size distribution dominated by coarse particles, with Angstrom exponent < 1 (e.g. Duvobik et al., 2002; Smirnov et al., 2003; Sayer et al., 2012a,b). However, as commented in the manuscript, from 30 September to 4 October 2011 (Fig. 4) columnar aerosol loads were high and were associated with relatively high values of α(440–870), which reached the highest value (about 1.6) on 4 October (indicating predominance of fine particles). During these days, the fine aerosol optical depth values were also high (> 0.19) and reached the highest mean daily value of 0.33 on 4 October. Thus, this behavior (high AOD and α(440–870)) is associated with the predominance of fine particles and not with strong marine aerosol contributions (as situations dominated by marine aerosol are characterized by low AOD and α(440–870)<1). In addition, the back trajectory analysis revealed that air masses during this event came from central Europe and passed over the Mediterranean Sea and the Spanish costal urban areas before reaching Alborán Island (Fig. 4c). Therefore, these air masses may have picked up fine anthropogenic particles emitted in these European urban areas before reaching Alborán Island, which may explain the behavior of AOD and α(440–870) during this event. For these reasons we think that this event is associated with aerosol anthropogenic transport from continental industrial/urban areas as there are no local anthropogenic activities at Alborán Island.

For these reasons, we decided to keep our discussion of Figure 4. However, to clarify this point in page 10 lines 19-23 we made the following change "Indeed, the back trajectory analysis revealed that this event was associated with air masses coming from central Europe and passing over the Mediterranean Sea and Spanish costal urban areas before reaching Alborán Island (Fig. 4c). Therefore, these air masses might pick up fine anthropogenic particles in their way to Alborán Island, which may explain the high values of both δ_a(500 nm) and α(440-870) parameters observed during this event."

Figure 5: Along with monthly mean values of δa, δF, δC , monthly mean values of alpha and FMF should be reported in another figure.

OK. We included figure5b representing monthly mean values of alpha and FMF in the new version of the manuscript.

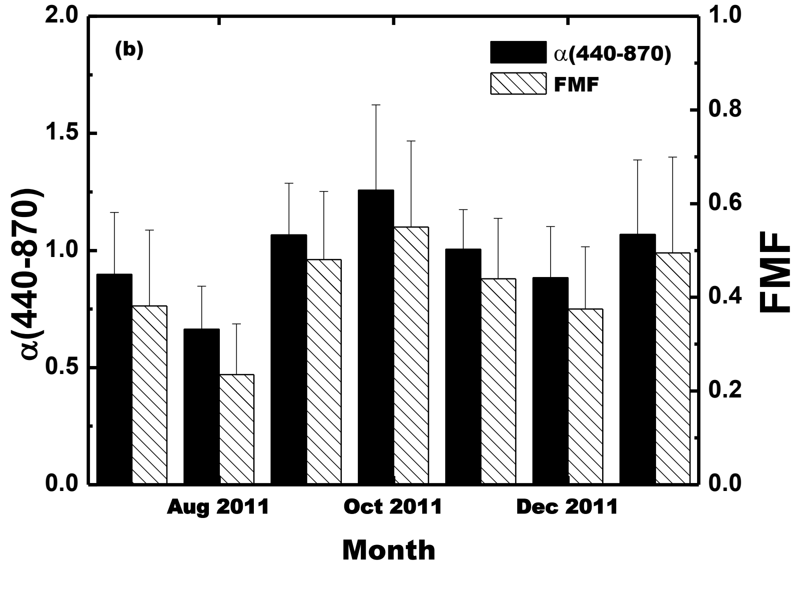


Figure 5b. Monthly variations of α(440-870) and fine mode fraction obtained at Alborán Island from July 2011 to January 2012.

Page 12: By the light of the previous considerations, authors should revise the comment on the figure 4c related to the pollution transported from Northern-Italy.

Please, see our previous response about this question.

Page 12: the strong reduction of the aerosol load during November-January could be related to the wet deposition too.

Ok. In the revised version, in page 11 lines 28-31 we made the following change "The low aerosol loads registered in November-January can be explained by the high frequency of clean Atlantic air advection (70-100%) and the absence of Saharan dust intrusions (Fig. 5c) as well as efficient wet removal aerosol processes due to cloudy conditions and precipitation in this period."

Page 13: For a better comprehension of the data analysis, authors should report, for each comparison, the number and the periods of coincident measurements days.

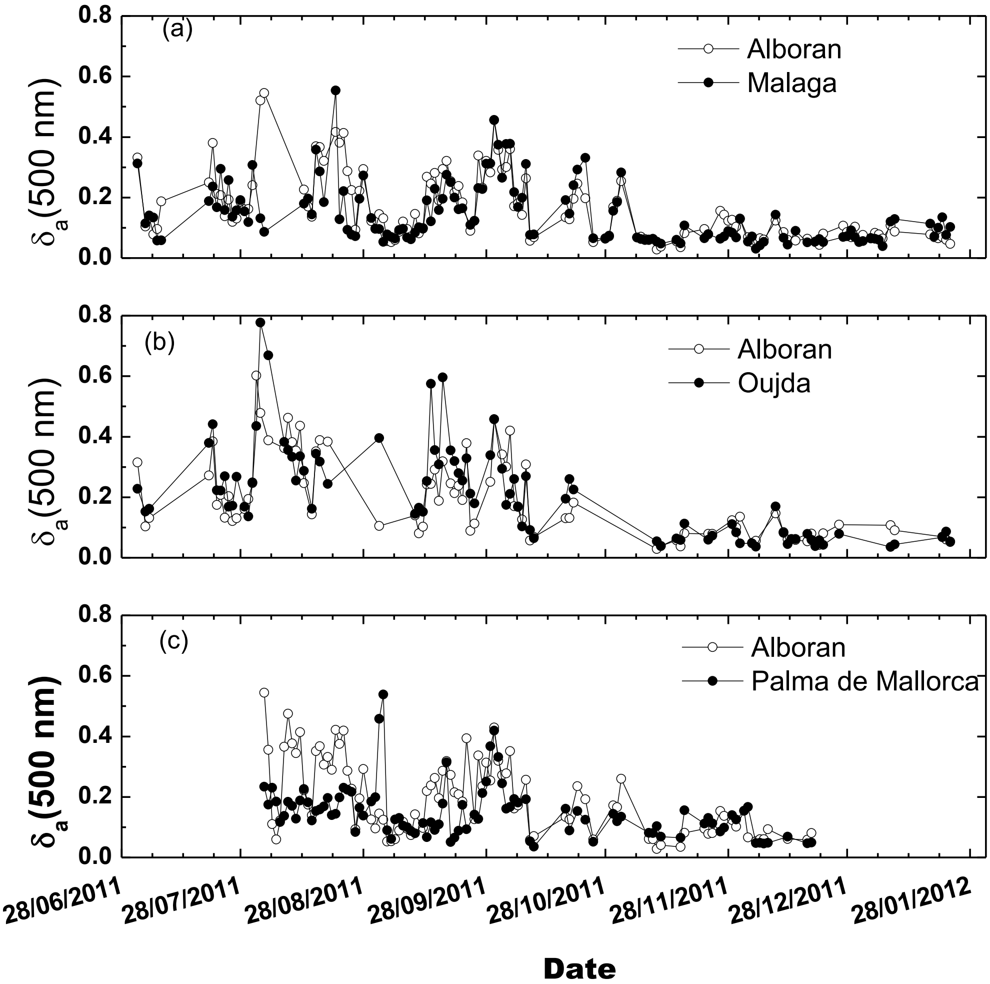
The numbers of coincident measurement days for each comparison were already reported in table 2 of the old version of manuscript. For clarity, in the revised version, in page 12 lines 27-31 we made the following change "Table 2 shows average values of $\delta_a(\lambda)$, α(440-870), δ_F(500 nm) and FMF as well as the number of measurement days for each comparison (Alborán – Málaga, Alborán – Oujda and Alborán - Palma de Mallorca). Only days with coincident measurements obtained at Alborán and at each one of the additional AERONET stations from 1 July 2011 to 23 January 2012 were used for direct comparisons."

Pages 13-14: Similar mean values of δF in Alboran and Malaga suggest similar concentrations of fine particles and, in particular, anthropogenic particles that are related to anthropogenic activities in Malaga and to ships emissions in Alboran. Did the authors verify the air masses paths for both sites in days with similar δF ?

For most of the days with similar δF, the air masses that affected both sites have almost similar paths. In general, the analysis of back trajectories for these cases revealed stagnation conditions over both sites.

It is recommended to show on different plots, as done in figure 6 for Malaga, the comparison between Alboran and Oujda and Alboran and Palma de Mallorca.

OK. We included the recommended plots (see below) in the new version of the manuscript.



The conclusion on the role of ships emissions on the Alboran atmosphere at the moment can be only an hypothesis. Analysis of radiometric data over more than one year (8 months) can better assess this. Anyway, only chemical analysis of particles sampled in-situ under different air masses circulation can unambiguously ascertain this.

We agree with the reviewer's that it is difficult to assess the effect of ship emissions using only radiometric data and that only chemical analysis can provide further insight about this effect. According to the reviewer´s comment in revised version, in page 13, lines 16-19, we changed our statement by "Thus, these results suggest that emissions from ships and/or from Mediterranean countries could play on Alborán Island a similar role that anthropogenic activities play in Málaga. Further studies using chemical analysis of particles sampled in-situ are needed to evaluate this hypothesis."

Page 15: Authors report the mean AOD(500) value for the entire cruise period (0.22 ± 0.12) that suggests a large aerosol load. This consideration is unimportant, since on table 3 mean AOD(500) values for each sector of the cruise depict different conditions, according to the crossed area (0.14 (East. Med ≤ AOD(500) ≤ 0.35 West Med.).

Ok. We removed the sentence "….that suggests a large aerosol load".

Moreover, the periods compared are different and aerosol loading can change also according to the seasons. The same consideration can be done for the other parameters. On the other hand, authors themselves highlight this on page 16, lines 9-11.

In this part of the manuscript we do not compare the results obtained in the different Mediterranean sectors. We only presented the data obtained during the cruise and those obtained for each sector and season (Fig. 7, Table3)

and discussed the possible cause of the observed aerosol variability during this cruise based on back trajectories analysis. However, we only compared aerosol loads obtained over the eastern Mediterranean during the cruise period 5-13 November and 18 August-13 September (Table 3) and we explained the observed drastic change in aerosol load from 5-13 November to 18 August-13 September by the seasonal changes in the meteorological conditions.

In any case, we have highlighted (as the reviewer commented) on the final part of this section that the spatial variations of aerosol properties over the cruise area implicitly includes the temporal variations of these properties. Thus, in order to avoid the inherent aerosol temporal variation in the cruise data, in the second part of this section we compared the cruise data for the different sectors with Alborán data (Table 4). In these comparisons we only used time coincident measurements.

For clarity in the revised version we separated this section in two sections: section "3.4. Variability of aerosol properties during MAN cruise" dealing with the aerosol variability during the cruise, and section "3.5. Comparison between Maritime Aerosol Network and Alborán measurements" where we compared MAN and Alborán measurements.

Page 15: On the basis of previous considerations, the comment on figure 4 is incorrect.

Please, see our previous response on figure 4.

Page 16: North-Eastern Europe is also an area strongly affected by biomass burning during summer.

Ok. In the revised version in page 15, line 15, we added the sentence "this region has been identified as a strong source of pollutants and biomass burning particles during summer"

Page 16: Authors explain the reduction of fine particles in Eastern Mediterranean from summer to autumn with the wet deposition and a less effective secondary aerosol formation. Wet deposition can be effective for large particles too.

We agree with the reviewer´s that wet deposition can be effective for large particles. However, according to Seinfeld and Pandis, (1998) wet deposition is more effective for fine particles than for large ones (Seinfeld and Pandis., 1998).

Seinfeld, J. H. and Pandis, S. N.: Atmospheric chemistry and physics: From air pollution to climate change, 1326 pp., John Wiley, New York, 1998.

Table 4: For a more complete information, authors are requested to report the number of coincident measurements Alboran/cruise sector for each comparison.

The number of coincident measurements Alboran/cruise sector for each comparison was already given in Table 4 in the old version of the manuscript. For clarity in page 16, line 20, we added the sentence "The number of coincident measurements for each comparison is also given in this table"

Page 17: Authors compare in table 4 the mean values of the radiometric parameters obtained in Alboran and in the different sectors of the cruise. They calculate these parameters grouping data from the same sector, although measurements were performed in different seasons. This could lead to uncorrect conclusions.

Following the reviewer criticism we compared the data obtained in Alboran and in the different sectors of the cruise for different seasons, using only time coincident measurements (see table4 below). It is important to note that, the number of coincident measurements Alboran/central Mediterranean sector for both 13-28 September

and 25 October-5 November periods and Alboran/central Mediterranean sector for 5-13 November period was small and thus these data were not used in the comparisons because they are not statistically significant. As can be seen the results in the table below are almost similar to those presented in Table 4 of the old manuscript. This table has been included in the revised version of the manuscript. In addition, in the revised version, in page 16, in lines 14-23 we made the following change "The number of coincident measurements obtained at Alboran and central Mediterranean sector for both 13-28 September and 25 October-5 November periods as well as those obtained at Alborán and eastern Mediterranean sector for 5-13 November period was small and thus the comparisons between measurements observed at Alborán and these sectors during these periods were excluded from this analysis. Table 4 shows the comparison between the mean values of δ_a(500 nm), δ_F(500 nm), δ_C(500 nm), α(440-870) and FMF obtained at Alborán and those obtained during ship cruise over Black Sea in the period 26 July-15 August as well as those obtained at Alborán and during ship cruise over the eastern Mediterranean Sea from 18 August to 12 September. The number of coincident measurements for each comparison is also given in this table"

	Alborán	Black Sea	Alborán	Eastern Mediterranean
δa(500 nm)	0.26±0.17	0.25±0.16	0.13±0.10	0.20±0.08
δ_F(500 nm)	0.08±0.04	0.21±0.14	0.07±0.04	0.16±0.07
δ_C(500 nm)	0.18±0.14	0.04±0.03	0.06±0.07	0.04±0.02
α(440-870)	0.5±0.4	1.7±0.3	1.0±0.3	1.7±0.2
FMF	0.37±0.17	0.82±0.08	0.55±0.12	0.81±0.08
Number of coincident measurements	26	26	28	28

Page 17: In a previous paragraph authors have strongly highlighted the similar contribution of fine particles to the AOD both in Malaga and Alboran: in one case they were mainly related to anthropogenic activities, in the other to the ships emissions. After the discussion in paragraph 3.4 one can derive that fine particles dominate Black Sea, central and eastern Mediterranean, unlike Alboran. This would mean that ship emissions in Alboran are not so important? In any case the possible ship emissions contribution in Alboran is lower than the contribution from anthropogenic emissions over Black Sea, central and eastern Mediterranean. Finally, are there information on the traffic of ships crossing Black Sea, central and eastern Mediterranean Sea and their related emissions?

In page 17 of the old version of the manuscript we have remarked that fine AOD (mean value of 0.09±0.06; see table2) in Alborán was surprisingly similar to the obtained in Malaga, Oujda and Palma de Mallorca, suggesting similar concentrations of fine particles over all these sites. As there are no local anthropogenic aerosol sources in Alboran (in contrast to Málaga, Oujda and Palma de Mallorca urban sites) we attributed this relatively high fine mode AOD at Alborán remote site to anthropogenic emissions from ships and/or urban-industrial area surrounding the Mediterranean Sea.

Later, in section 3.4 we have highlighted that fine particles dominate Black Sea, central and eastern Mediterranean, and that fine mode AOD obtained over these sectors was higher than the obtained over Alboran (Table 4), which we attributed to the large contribution of anthropogenic emissions over Black Sea, central and eastern Mediterranean in comparison to Alboran.

Shortly, in this work we highlighted that the fine aerosol load in Alborán Island was similar to the obtained in Málaga, Oujda and Palma de Mallorca urban sites, which suggest a significant anthropogenic emission influences (comparable to anthropogenic emission influences over Málaga, Oujda and Palma de Mallorca urban sites) over Alboran remote site. On the other hand, we also highlighted that Black Sea and eastern Mediterranean sectors are more influenced by anthropogenic emissions than Alborán Island (western Mediterranean) as indicated by the high fine aerosol loads in Black Sea and eastern Mediterranean sectors in comparison with alborán.

In any case, in order to avoid confusion we separated the section 3.4 in two sections: section "3.4. Variability of aerosol properties during MAN cruise" dealing with the aerosol variability during the cruise and section "3.5. Comparison between Maritime Aerosol Network and Alborán measurements" where we compared MAN and Alborán measurements.

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

The paragraph on the effects of the EU regulations on the air quality in Malaga seems very untied from the rest of the paper and should be removed. In fact, radiometric data only are not enough to evaluate aerosol loading dynamics at the ground. This can be an argument for a different paper, supposed that measurements from ground-based instruments are analyzed together with columnar ones.

Ok. We removed this section.

Page 19: Authors should revise the percentage of measurements in background conditions, according to the previous observations on what are the background conditions.

See our previous response about this point.

Page 19: The conclusion "The mean value of τ_F(500 nm) over Alborán Island was comparable to that observed over the other three nearby AERONET stations, suggesting homogeneous spatial distribution of fine particle loads over the four studied sites in spite of the large differences in local sources." should be verified because, at the moment, the reader does not know how many coincident measurements have been considered for the comparison of each measurements site.

The number of coincident measurements was reported in Table 2 (see our previous response).

Finally, all the conclusions should be revised according to the revisions required.

OK

Minor revisions

Figure 2a: For sake of clearness it would be better to report mean AOD values at two wavelengths.

OK

Page 5, Line 12: Authors have already said that Western Mediterranean area is poor of aerosol measurements.

OK. We removed this sentence

Geographical coordinates of Alboran in tnhe abstract are different from those used for back-trajectories calculation and reported in the corresponding plots.

Ok. We corrected this mistake

Page 14: It is unnecessary to repeat that the comparison between Alboran and Oujda was done with coincident measurements.

OK. We removed this sentence

Page 15: change Figure 7c in Figure 7b.

Page 17, line 3: change “lower” with “higher”.

Page 17, line 6: delete “significantly”.

Page 17, line 18: change “can explains” in “can explain”.

Ok. We corrected these mistakes