Interactive comment on "Continental pollution in the Western Mediterranean Basin: vertical profiles of aerosol and trace gases measured over the sea during TRAQA 2012 and SAFMED 2013" by C. Di Biagio et al.

At first, we would like to thank the reviewer for having carefully read the paper and provided valuable comments which helped to improve the quality of the manuscript. We have taken into consideration all the questions raised by the reviewer, and changed the paper accordingly. The details of our changes are highlighted in the text. The point by point answers to Reviewer #1 are provided in the following.

Anonymous Referee #1
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The paper focuses on the analysis of aerosol and trace gas vertical profiles obtained over the sea in the Western Mediterranean Basin during the TRAQA 2012 and SAFMED 2013 summer campaigns. Even though the number of measurements presented in this paper is relatively short (23 profiles), it reasonably covers large area in the western Mediterranean basin providing insight of the impact of the different pollution transport regimes.
I have some suggestions for a minor revision of the manuscript (see below).

Specific comments

Abstract Lines 8-10 page 8285: this sentence is unclear. Maybe the authors mean that during TRAQA and SAFMED campaigns the study area was under a wide range of meteorological conditions that favored the pollution export from different sources located around the basin which allowed sampling atmospheric aerosols of different origin and types. Please, rephrase this sentence.
The abstract has been corrected as suggested by the reviewer.

Lines 13-15 page 8285: authors state that aerosol layers not specifically linked with Saharan dust outflows are distributed ubiquitously which indicates "quite elevated levels "of background pollution throughout the Western Basin. This statement is not justified by data analysis presented in this paper. Authors do not presented the analysis of background conditions over the study area. Please clarify this point and provide information that can justify these "quite elevated levels "of "background pollution".
The sentence "Aerosol layers not specifically linked with Saharan dust outflows are distributed ubiquitously which indicates quite elevated levels of pollution throughout the Western basin" has been eliminated from the abstract. We agree that it is not precise.

Introduction The recently published papers by Valenzuela et al., 2014 and Lyamani et al., 2005 should be referred in this manuscript. Valenzuela et al., 2014. Aerosol transport over the western Mediterranean basin: Evidence of the contribution of fine particles to desert dust plumes over Alborán Island. Journal of Geophysical Research D: Atmospheres, 119 (24), pp. 14028-14044 Lyamani et al., 2015. Aerosol properties over the western Mediterranean basin: Temporal and spatial variability. Atmospheric Chemistry and Physics, 15 (5), pp. 2473-2486.
Despite their interesting results, these papers analyses data from a region quite far to our investigation area and much more under the influence of different meteorological/aerosol

export conditions compared to those analysed in this study. For these reasons we have not included these two valuable papers in the references.

Lines 2-3 page 8288: Please correct by “During TRAQA and SAFMED the Western Basin was under diverse synoptic conditions”.
The correction has been made.

Line 4 page 8288: Please provide a brief description of Mistral/Tramontane events. This will help understand the interpretations of the results.
We have added a brief description of the Mistral/Tramontane in Section 4, when describing the different meteorological conditions: “On the same days a strong Mistral-Tramontane episode (i.e., strong northerly winds developing along the Rhône and Aude valley which bring a northerly/north-westerly flow over the Western Mediterranean, see Fig 3a) favoured the dispersion of pollutants towards the central part of the Western basin”.

Lines 7-8 page 8288: The authors state that the main objective of the present work is to provide “extensive observations” of the vertical distribution of aerosols and trace gases. However, they only present 23 profiles. Please, be precise.
The term “extensive” has been eliminated.

Section 3.2 Line 1 page 8293: between 0.1 and 3.0 _m or between 0.11 and 4.17 _m? Please check.
The text is correct and is well 0.1-3.0 µm; however we found a typos in page 8290 line 12 since the CPC range is 0.004-3.0 µm, instead of 0.004-1.0 µm as stated in the text. This has been corrected in the text.

Line 19 page 8294: eq. (2) instead eq. (1)
The correction has been made.

Section4. A brief description of the AERONET data (data level, accuracy of the data, etc.) and the instrument used should be provided. In addition, information on AERONET sites (characteristic, location, etc.) should be given. Please include these sites in Fig.1. This may help to make clear the interpretation of the results.
The first paragraph of Section 4 has been rewritten as: “In order to characterize the general aerosol conditions encountered over the Western Mediterranean basin during the TRAQA and the SAFMED campaigns we have plotted the time-series of the aerosol optical depth (τ, ±0.02) at 440 nm and the 440-870 nm Ángström exponent (α) measured with a Cimel sunphotometer (Holben et al., 1998) at the three AERONET stations of Barcelona, Frioul, and Ersa located along the coast around the Western basin (see Fig. 1). Level 1.5 cloud-screened data are used in this study.”
Fig. 1 has been also modified to include the 3 AERONET stations.
For the AERONET data I used the Level 1.5 for which data from all the stations were available. For the stations for which they were available, the comparison with Level 2.0 data does not indicate any difference in the interpretation of the results.

Lines 19-20 page 8295: it is obvious that TRAQA campaign in 2012 was characterized by very variable meteorological conditions than SAFMED campaign because TRAQA campaign period (20 June–13 July 2012) was larger than SAFMED campaign period (24 July–1 August 2013). On the other hand, I don’t understand how very variable meteorological conditions can prevent the accumulation of high levels of pollutants over the basin. Some meteorological

conditions as discussed later by the authors were responsible of high pollution events during TRAQA campaign. Please clarify and rephrase this sentence.
The sentence "The TRAQA campaign in 2012 was characterized by very variable meteorological conditions which prevented the accumulation of high levels of pollutants over the basin" was probably speculative. The whole sentence has been eliminated from the text.

The title ("Events observed") of the last column of table 1 is not adequate. For example "Test flight" , "Follow of Barcelona pollution plumes " and "Characterization of pollution in central Italy" are activities that have been carried out and not events that were observed during these campaigns. Please correct.
The column title has been changed in "Description".

Line 15 page 8295: authors state that Fig. 2 show data corresponding to the period of the campaign of measurements plus 10 days before and after. However, Barcelona data (left panels) correspond to the period of the campaign plus 1 day before and after. Please check.
Data for the Barcelona station are not available over the whole considered period in 2012. The caption of Figure 2 has been rewritten as: "Aerosol optical depth at 440 nm (τ) and Ångström exponent (α) measured at the Barcelona, Frioul, and Ersa AERONET stations during the TRAQA 2012 (left panels) and the SAFMED 2013 (right panels) campaigns. The time period for the different plots is ±10 days around the beginning/end of the two campaigns (data for the Barcelona station are not available over the entire period for 2012). The label D indicates the days affected by Saharan dust".

Lines 21-22 page 8295: the aerosol optical depth was below 0.2 before the beginning of the campaign over the three analyzed AERONET sites and not over the whole basin. Please be precise. In addition, authors sate that the aerosol optical depth (AOD) increased to 0.3–0.5 (with 1 < alfa< 2) in the periods 23–26 June and 3–13 July. However, as can be seen in Fig. 2 the aerosol optical depth was in general below 0.2 over the three AERONET sites during 23–26 June. Also, the AOD was in general below 0.3 (especially at Frioul and Ersa) during 3–13 July. However, from fig2, it can be seen that the aerosol optical depth was relatively high from 30 June to 4 July which was associated with dust intrusions. Please check and correct.
The sentence has been rewritten as: "Over the analysed AERONET sites the aerosol optical depth was below 0.2 before the beginning of the campaign and increased, especially at Barcelona and Ersa, to ~0.3-0.5 (with 1<α<2) in the periods 23-26 June and 3-13 July".

For clarity please reduce the y-scale of fig.2.
The y-scales for the optical depth and Angstrom exponent have been reduced as much as possible.

The authors present Fig 3, but, don't discuss it. Please, discuss this figure or remove it from the paper because it does not add any significant information.
Figure 3 can be useful to identify the different meteorological conditions encountered during the campaign, as for example the Mistral/Tramontane event cited by the reviewer, so we decided to maintain this Figure. Reference to Fig. 3a and 3b has been made within the text of Section 4.

Lines 4-6 page 8296: Please, specify if this export regime has occurred at all altitude levels or at specific altitude. The same should be done for the other described export regimes. This is because export regime can be different at different altitudes.
The main objective in Section 4 is to provide an overview of the main different conditions encountered during the different flights, so to understand the general strategy and observed continental outflow regimes. A more detailed analysis of the different flights, including wind vertical profiles and back-trajectory analysis, is performed in Sect. 5. We do not think necessary to specify all the details here.

Lines 4-6 page 8296: authors state that on 26–27 June north/north-easterly winds blew across northern Italy determining an air mass outflow towards the Gulf of Genoa. However, Fig.9 shows that the air masses on 27 June come from France and not pass over Italy. Please check and correct.
We have checked and the text has been changed in north/north-westerly.

Lines 3-19 page 8296: Mediterranean Sea and ship traffic are important source contributing to the aerosol loading over the Mediterranean Sea. The authors cannot a-priori exclude this important sources.
Ship emissions are undoubtedly a very important source of pollution over the sea mostly close to the surface. During TRAQA and SAFMED flights covered an extended altitude range up to 5000-6000 m, so ship emissions can influence the profiles predominantly in the lowest atmospheric layers. A reference to the possible impact of ship emissions has been added in Sect. 5.4 in reference to flight V28 discussed there.

Line 12 page 8296. I think that authors refer to Fig 1 and not to Fig3. Please verify.
The correction has been made.

Lines 12-14 page 8296: this sentence seem to be not related to this section. To be consistent, authors should describe the meteorological/export during flight V31 as they did for the other flights. Please, explain what you mean with “moderate” Mistral episodes.
For V31 the sentence has been rewritten as: “Additionally, flight V31 sounded the atmospheric structure close to the Spanish coasts reaching the southern urban area of Valencia. The flight was performed under the influence of south-westerly winds favouring the export from the Iberian Peninsula towards the basin”.
The reviewer is correct and “moderate” is not necessary, so I rewrote as “(iv) Mistral episodes occurred on the 6-7 and 11 July 2012”.

5 Results Figure 5 is poorly discussed and interpreted. More discussion and interpretation of the results presented in this figure is needed. If no more discussion and interpretation of this figure is given this figure should be removed from the paper.
I understand that the reviewer does not consider Fig. 5 sufficiently discussed, however it represents the starting point of whole Section 5. The fact observing such variability in the scattering coefficient, particle concentration, and trace gases as shown in the figure is the key point that motivates the detailed investigation of the different flight observations. Moreover, Fig. 5 summarizes the range of observations encountered during the campaigns, so providing a reference for pollution conditions in the Western Mediterranean basin.

Section 5.1 The figures presented in fig.6 should have the same X and Y scales. Also, vertical profile of Angstrom exponent should be included in this figure. This in combination with dNAcc and dNcoarse will help to identify the type of aerosols dominant in each layer.

The x and y scales in Fig. 6 have been corrected. Conversely, for what concerns the Angstrom exponent, we have decided not to add it to the plot. We find that it does not add any further information to the plot. In fact, the information on the type of the particles is already given by the spectral variability of the scattering coefficient (pronounced spectral variability corresponds to small particles, so to an higher Angstrom exponent; a neutral spectral variability corresponds to large particles, so to a lower Angstrom exponent).
Finally, Fig. 6 already contains a large number of information and adding another parameter would reduce the clarity of the figure.

Scattering profiles during TRAQA campaign (6 s resolution) are nosier than those observed during SAFMED (1 s resolution). Please give an explanation for this.
The stronger noise during TRAQA is probably due to the non-perfect cleaning of the nephelometer cavity during this campaign compared to SAFMED, for which the nephelometer was cleaned before the beginning of the campaign. This effect however does not affect the interpretation of the results.

Lines 8-10 page 8298: authors state that the profile of the aerosol scattering coefficient is mostly correlated to dNAcc., however, they not justify this statement. Please give statistical parameter that justifies this statement.
What we wanted to highlight in this section was the link between the particle concentration in the accumulation mode and the scattering coefficient, which is quite evident by looking at the plots, and not to estimate quantitatively this relation. In order to avoid any confusion on this point the text has been changed in "The structure in the scattering profile is generally mirrored in dN_{Acc} profile,".

Lines 16-20 page 8298: Please, provide an explanation of the cause of scattering coefficient and dNAcc maxima and minima.
As stated in the text the minima and maxima during SAFMED are associated to the different phases of the campaign, the first more polluted and the second one less polluted as discussed in Sect. 4. Concerning TRAQA, the absolute maxima are obtained in correspondence of the dust event. Some details have been added in the text.

Lines 3-5 page 8299: authors state that the scattering coefficient and the particle concentration measured in the FT are comparable with the values observed in the BL, and in few cases even larger (V25, V26, V30). This contradicts with the results shown in fig. 5. Please check and correct.
This sentence refers to the fact that in single profiles the measured values of the scattering coefficient and particle concentration may be larger in the FT than in the BL, as it is observed in some profiles (see Fig. 6). The box and whisker plot, conversely, describe the entire dataset and gives an overview over all conditions. This point has been specified in the first paragraph of the Results Section.

Lines 16-19 page 8299: Please, include Table comparing your results with those found in literature. Also, for better comparison (same experimental set), authors should include their results obtained over land in this table (e.g. flight V49).
A new table (Table 2) has been added. This table resumes the measurements of dN_{Aitken} and dN_{Acc} in the BL and in the FT for the entire set of observations, also in comparison with literature data obtained over continental Europe. Data for V49 over land during SAFMED has not been added to the Table since no data are available for the CPC for the majority of this flight, so no information on dN_{Aitken} can be retrieved.

Lines 16-19 page 8299: during flights V52, the scattering coefficient was very low and no pollution aerosol layer can be seen in this flight as confirmed by authors. Authors should include a Table comparing the data obtained under the different main meteorological/export conditions. This will help to identify the main cause (and sources) of high pollution levels over Mediterranean Sea.

For TRAQA and SAFMED we measured different outflow conditions which can be useful to describe the complexity of the export towards the basin. However comparing data for the different cases would appear quite complicated. We consider that adding such a Table, as suggested by the reviewer, would require having a larger statistic of cases.

Section 5.2 Lines 26-28 page 8300: please give references that support your statement. Also, include Table comparing your results with those obtained during flight over land and with those found in literature.

As a support to my statement I have added the reference by Parrish et al. (1998) which define moderate pollution when CO<180 ppbv. CO and O_3 have not been added in Table 2, however we have rewritten the first paragraph of Section 5.2 to take into account your suggestion: "CO and O_3 vary in the range 60-165 ppbv and 30-85 ppbv, respectively. The 25^{th} and 75^{th} percentiles are 87 and 105 ppbv for CO and 49 and 62 ppbv for O_3, representative of moderate pollution conditions (Parrish et al., 1998). By comparison, the values measured over land in central Italy during flight V49 are in the range 80-180 ppbv for carbone monoxide and 40-85 ppbv for ozone".

Section 5.3 Lines 11-13 page 8302: Please provide Table comparing your results with those reported by these authors.

We have added the values of the reference papers cited in parenthesis within the text of Section 5.3. The text has been rewritten as: "respectively, with a corresponding $\Delta O_3/\Delta CO$ ratio which varies in the range ~0.10-2.0 for all cases. These values are comparable with the range of observations available in the literature for fresh and moderately aged pollution plumes in the BL and in the lower FT (~0.2-1.0) (Chin et al., 1994; Parrish et al., 1998; Zhang et al., 2006; Cristofanelli et al., 2013)".

Section 5.4. Line 24 page 8302: statistical analysis should be provided to justify the good correlation between dNAitken and dNAcc, CO, and O3. From Fig. 10 it can be seen that dNAitken and dNAcc, CO, and O3 are not correlated.

The text has been corrected and rewritten as: "For about half of the observed events the dN_{Aitken} layer appears related to a simultaneous increase in dN_{Acc}, CO, and O_3, which suggests that the layer has been transported from a region directly emitting in this size range".

Lines 7-24 page 8305: I think that this dNAitken event can be simply associated to ship emissions.

We have rewritten the sentence as: "For the V28 layer (Fig. 10b) the dN_{Aitken} is correlated with CO which might indicate the influence of local emissions close to the surface level (i.e., ship emissions)".

Conclusion

Lines 19-21 page 8307: authors state that the geographical distribution of aerosols and trace gases observed in this study appears quite homogeneous within the investigated area, suggesting a relatively similar contribution from the various sources located around the north-

western basin. However, the results presented in this paper show that the aerosol and gas profiles obtained in different areas in the Mediterranean basin show very different stricture and composition. Please clarify this point.

We agree with the reviewer and the entire sentence has been eliminated from the text.