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 #2 are provided in the following.

Anonymous Referee #2

The manuscript presents airborne measurements of aerosols and trace gases CO and ozone over the Mediterranean focusing on the vertical distribution of several compounds within a series of vertical profiles between Spain, Corsica and the Gulf of Genova. Ozone and CO values respectively their ratio are used to characterize air masses. Such vertical distribution data over the Mediterranean are very scarce. They show that both Saharan dust and continental pollution are present in large amounts. Several of the compounds measured are not detectable from the ground or from remote sensing techniques although they are possibly crucial for the Mediterranean climate. It is highly recommended that such data are getting available.
However, the manuscript has several weaknesses that need some further work especially in the detailed description of the individual profiles. Generally the graphics of Fig. 6, 8 and 10 lack the size and resolution required. Within the text often a series of profiles are mentioned. For the reader it's difficult to find these profile data without having an indication in which of the different figures these data are contained. Some figures are labelled a or b without having a description in the text. For example V28 in Fig. 10 looks different from V28b in Fig. 6.

Following the reviewer suggestion we have tried to clarify the references to the Figures where it seemed confusing. Concerning Fig. 6, 8, and 10 they can be reproduced with a larger size in the published version of the paper. This will considerably help their clarity and quality.
The same profile (V28b) is shown in Fig. 6 (between 0 and 4000 m) and Fig. 10 (between 0 and 1500 m). V28 has been corrected in V28b in Fig. 10.

Specific comments:

Page 8292, section 3.2.: There is a bit of confusion about OPC and PCASP measurements. If another OPC (GRIMM) is onboard, avoid OPC for the PCASP.
The term "OPC" has been removed from the text and replaced with PCASP.

Page 8293, STP Conversion: line 7 and 8, ozone is measured using UV absorption. This technique is pressure dependent and pressure has to be taken into account. Does the MOZART instrument correct for pressure over all the altitude ranges?
The MOZART instrument itself does not correct pressure but it is connected to a pressurized inlet on-board the aircraft which compensates outside pressure drop during the flight. The

pressure inside the inlet is maintained constant (and monitored) at 1020 hPa during the whole flight.

Page 8296, line 4-6, The text claims northeasterly winds, the data in Fig. 8 show northwesterly winds in altitudes above 1500 m.
The reviewer is correct and we stated north/north-easterly in spite of north/north-westerly, as shown both in Fig. 3 and 8. This error has been corrected in the text and in Table 1.

Page 8298, line 10: typical of pollution/anthropogenic particles. . . needs more description what is the 'typical spectral variability
The sentence has been rewritten as: "For the different vertical soundings the particle concentrations dN_{Acc} and dN_{Coarse} vary in the range ~100-3000 scm^{-3} and ~5-4000 scm^{-3}, respectively, for plumes with σ_s between 10 and 120 Mm^{-1}. The profile of the aerosol scattering coefficient is mostly correlated to dN_{Acc}, and this also reflects the pronounced spectral variability (i.e., decrease for increasing wavelength) of the scattering coefficient, typical of pollution/anthropogenic particles. dN_{Coarse} also contributes to the scattering signal in some cases especially at high altitudes (see V16, V20, V21, V22, and V23 above ~2000 m), and this reflects the low spectral variability of the scattering coefficient".

Page 8299, lines 18 and 19: The manuscript states that these values are comparable with values measured close to the surface at urban continental sites but the references are taken from rural (Petzold), airborne (Mallet 2005), the proper reference is Mallet 2003, rural to suburban (Wiegner instead of Weigner) rural Po-Valley (Junkermann) airborne, (Hamburger).
Following the reviewer suggestion we have rewritten as: "The dN_{Acc} and dN_{Aitken} measurements within the BL and in the FT over the sea are comparable with the values measured close to the surface at continental sites under pollution conditions (Petzold et al., 2002; Mallet et al., 2003 and 2005; Wiegner et al., 2006; Junkermann, 2009; Hamburger et al., 2012)." Concerning the reference of Mallet et al., the papers of 2003 and 2005 analyses data acquired during the same campaign (ESCOMPTE in 2001) over the area of Marseille/Fos Berre. I have however added the Mallet et al. 2003 reference to the list.
I have also added the following reference to the comparison:
Highwood, E. J., Northway, M. J., McMeeking, G. R., Morgan, W. T., Liu, D., Osborne, S., Bower, K., Coe, H., Ryder, C., and Williams, P.: Aerosol scattering and absorption during the EUCAARI-LONGREX flights of the Facility for Airborne Atmospheric Measurements (FAAM) BAe-146: can measurements and models agree?, Atmos. Chem. Phys., 12, 7251-7267, doi:10.5194/acp-12-7251-2012, 2012.

Page 8300, section trace gas vertical profiles: The authors discuss ozone in freshly polluted and aged air masses. In freshly polluted air masses ozone is normally titrated with coemitted nitrogen oxide. In this case a peak in pollution (without other parameters probably here CO) would be visible together with a reduction of ozone in the same layer, see Figure 10. However, in the vertical profiles these features are not coincident. The particle peak is lower in altitude than the ozone dip. That's looking like a mismatch of the timing in the data. Very similar in V20 the clean layer in the scattering data are between 1600 and 200 m. the concurrent ozone peak is about 100 m lower. Such a timing mismatch can have consequences for the ozone / CO ratio which is used for further analysis.
We have verified and there are not mismatches between ozone and the other data shown in Fig. 8 and 10. In the different profiles O_3 appears mostly correlated with dN_{Acc} and vertical localization of the peaks is mostly coincident. However it should be taken in mind that the

processes controlling particle concentration/optical properties and the gas chemistry may be not the same, and this can influence their vertical distribution.
Concerning the expected minimum in ozone in correspondence of fresh pollution plumes, it has to be pointed out that measurements are performed over the sea and not close to continental/urban sources, so it might be expected that the O_3 concentration within plumes can vary depending on the photochemical processes and on possible mixing occurring along pollution export over the sea, as well as the concentration of NO_x and VOC (Volatile Organic Compounds) at emission and their time evolution.
The estimation of the titration, in any case has not been possible during TRAQA when NO_x were not measured. During SAFMED, NO_x were measured and we observed titration of O_3 only in one case very locally above the sea surface (at about 150 m) in the gulf of Genoa during flight 51 (data in correspondence of a straight levelled run, not shown in this paper). This was possibly linked to fresh ship emissions. No other cases of O_3 titration were observed during SAFMED.

Page 8302, section 5.3. For the O3 and CO ratios 'typical' values are given. This is not the case for the Aitken to accumulation number ratio. It would be good to have some idea about such 'typical' values.
We did not found in the literature some reference values for the Aitken to Accumulation particle mode ratio for pollution particles. We have found several references, but mostly for forest fires aerosols. If the reviewer has some references to suggest we would add it in the paper.

Page 8303, Section 5.3.1, Profile V19: Contrary to the text the profile shows values of about 3000 Aitken particles up to about 1500 m. the lowest values were measured just above, not below 800 m. This is just above the MBL as indicated and shows a peak in the accumulation mode and in the humidity. The ratio of Aitken to accumulation mode particles rises rapidly above 2500 m. This is not discussed in this section at all. It's mentioned a bit later in the text, but should be included here. Again there is an altitude mismatch between observations of increased Aitken mode particles that are described as fresh emissions and the concurrent ozone measurements.
The distinction of the two layers (the one below 800 m and the second one at 800-2600 m) has been performed based on the dN_{Aitken}/dN_{Acc} and $\Delta O_3/\Delta CO$ profiles, which show a distinct behaviour at the two considered altitude ranges. The single profile of dN_{Aitken} does not permit to distinguish these two structures. We changed the text accordingly to clarify this point in the text. Additionally, we also mention here the presence of the layer above 2600 m characterized by high dN_{Aitken}.
Concerning the possible altitude mismatch between dN_{Aitken} and O_3, see the answer to one of the previous comments.

Page 8304, Section 5.3.2 V20 The CO rich layer is only within the lowest 150 m. no data are presented in Fig. 8. Data in the figure are not always in agreement with the text. Lowest Aitken number concentrations at 380 m are very low, clearly below 1000 in the same altitude also the scattering coefficients are typical for the free troposphere.
The text has been changed as: "The aerosol profile in the BL is characterized in the first ~400 m by the presence of a layer richer in dN_{Aitken} ($dN_{Aitken}/dN_{Acc}>20$) and CO (100 ppbv close to the surface; CO data not available between 150 and 650 m) possibly linked to fresh pollution, followed by the alternation of several layers characterized by a variable dN_{Aitken} (1000-6000 scm^{-3}) and lower CO (~70 ppbv). A local minimum of dN_{Aitken} and σ_s is found at ~400 m."

Page 8306, Line 22 ff. The initial text of the paragraph is confusing, first of all case studies are mentioned in Fig. 10, than several profiles are listed, but finally only two of those are included in the figures.

I guess the reviewer refers to page 8005. The initial text of the paragraph has been rewritten as: "For about half of the observed events the dN_{Aitken} layer presents a good correlation with dN_{Acc}, CO, and O_3 which suggests that the layer has been transported from a region directly emitting in this size range. These cases are: V16 at ~200-400 m, V21 at ~400-800 m, V28 at ~250 m, and V31 at ~1000-3000 m (only V28 and V31 are shown in Fig. 10).".

Pages 8305/8306, High Aitken number concentrations are described as originating from Valencia. That would require a very intense vertical mixing up to 3000 m. What is the reason for the low values in the marine Boundary layer, despite rather high values of CO (Fig. 10)?

The back-trajectories analysis indicates that in the boundary layer the air-masses have a different origin compared to the free troposphere. In particular, they come from the open sea (eastern of Valencia) in the boundary layer. This can explain the different behaviour observed in the V31 profile. This point is specified in the text.

For reference, several papers have focussed on the export mechanisms in the Western Mediterranean basin within the lower troposphere, such as:

Millán, M. M., B. Artíñano, L. Alonso, M. Navazo, and M. Castro: The effect of meso-scale flows on the regional and long-range atmospheric transport in the western Mediterranean area, Atmos. Environ., 25A, 949–963, 1991.

Velchev, K., Cavalli, F., Hjorth, J., Marmer, E., Vignati, E., Dentener, F., and Raes, F.: Ozone over the Western Mediterranean Sea – results from two years of shipborne measurements, Atmos. Chem. Phys., 11, 675-688, doi:10.5194/acp-11-675-2011, 2011.