

Interactive comment on “Primary marine aerosol emissions from the Mediterranean Sea during pre-bloom and oligotrophic conditions: correlations to seawater chlorophyll a from a mesocosm study” by A. N. Schwier et al.

We thank the reviewer for their helpful comments and discussion. The concerns of this reviewer are addressed below.

Major comments: - Investigating the impact of ocean acidification on primary marine organic aerosol emissions is one of the major goals of this paper. However, it is not clear for me that simply adding CO₂ in marine water during few days can mimic properly the impact of future acidification on marine aerosol emission. I assume that future climate changes may, on a long term run, affect organic content in seawater (phytoplankton species and concentration, virus population . . .) in the Mediterranean. Also, climate change may not only affect pCO₂ but also wind speed which is an important parameter in marine aerosol formation (Bopp et al., 2001; 2003). At least few sentences should be added in the manuscript to describe these issues.

We thank the reviewer for pointing this out. We answer their comment below and in the following comment response. Indeed, our experiment was only focused on the effects of ocean acidification on the composition and function of Mediterranean plankton communities, without taking into consideration the potential effects of climate change related processes such as warming and modifications of wind regimes. These parameters may also change, but were outside the scope of this study. It is important to keep in mind the idea that planktonic organisms have a generation time of a few hours to a few days, and that as such they can react very quickly to an external driver, such as adding CO₂ in a mesocosm. Many experiments, with some following the same protocol as in this work (mesocosms deployed for multiple days), have already shown that increased CO₂ availability could lead to important modifications of planktonic community compositions and functioning. For more explanation, we invite the reviewer to reference (Gazeau et al., submitted) for more details of these campaigns and the reasoning behind them.

We have modified our introduction to develop these considerations and to better explain why ocean acidification would potentially affect marine aerosol formation, through the modification of plankton community composition and organic matter production. We have added the following paragraph into the introduction to more fully explain this: “The production of organic matter in oceanic surface water is expected to be substantially modified in the coming decades as a consequence of climate change and ocean acidification (Doney et al., 2012). Ocean acidification is defined as the increase in ocean acidity and associated changes in seawater chemistry, due to the absorbance of a very significant amount of anthropogenic CO₂ by the oceans (2.5 ± 0.5 Gt C or ~26.3% of anthropogenic emissions, Le Quéré et al., 2014). Since the beginning of the industrial era, the pH in ocean surface waters has already decreased by 0.1

units, on average, equivalent to an increased acidity of 26%. Further acidification is expected by 2100, ranging from 0.06 to 0.32 units, equivalent to an increased acidity of 15 to 110%, depending on the considered CO₂ emission scenario (Ciais et al., 2013). Although it is well established that ocean acidification has the potential to significantly impact marine biological processes (see Riebesell and Tortell (2011) and Weinbauer et al. (2011) for a comprehensive review), it is still unclear how these changing biogeochemical water conditions will affect the properties and production of marine aerosols. Furthermore, the effect of such an acidification and consequently the resulting feedback on Mediterranean marine aerosol and the regional climate remains unknown.”

We have added the following statement to the conclusions to further expand this point, “It is important to note that there are additional effects, such as wind speed, precipitation levels, and temperatures, that could change with future climate change and that these were not included within this analysis; instead, we focused on ocean acidification effects on Mediterranean Sea plankton communities and subsequent effects on primary marine aerosol. Future studies will need to incorporate additional parameters to determine further effects on primary marine aerosol.”

Why should we investigate ocean acidification specifically in the Mediterranean? This should be better justified (in few sentences). “Mediterranean marine aerosol remains relatively uncharacterized”: this may be not enough to justify the need for characterizing them.

We have modified the introduction to include the following information to describe in more detail the importance of studying acidification experiments of Mediterranean aerosol, “Mesocosms are defined as experimental enclosures from one to several thousands of litres that maintain natural communities under close-to-natural conditions (Riebesell et al., 2013). They have been increasingly used in both aquatic and terrestrial ecology (Stewart et al., 2013), especially on the effects of environmental and/or anthropogenic disturbances on a large variety of chemical and biological processes. In the context of ocean acidification, mesocosms have been used on several occasions for experimental time periods spanning from a few days to a few weeks, and were found to be efficient in studying the effects of this driver over such short time scales (Riebesell et al., 2008, 2013). Archer et al. (2013) recently showed, during a mesocosm experiment in the Arctic, that with seawater acidification and increased CO₂ concentrations, average concentrations of DMS decreased by up to 60% at the lowest pH. Inversely, concentrations of DMSP, the precursor to DMS, increased by up to 50%. In the remote ocean, DMS was predicted by modeling studies to be one of the main precursors for CCN in the marine boundary layer, and studies have shown that regional DMS emission changes could affect CCN sensitivity (Cameron-Smith et al., 2011; Woodhouse et al., 2013).

Many past mesocosm experiments which focused on the effects of ocean acidification have been performed in relatively eutrophic conditions or with nutrient addition initially or during the

experiment. However, about 60% of the ocean surface is associated with low productivity, termed oligotrophic areas. Decreased nutrient availability and the expansion of low productivity regions are projected with increasing CO₂ concentrations, as enhanced thermal stratification is expected to lead to surface layer nutrient depletion (Irwin and Oliver, 2009; Polovina et al., 2008). Nutrient availability also might have strong effects on the community response to ocean acidification (Hare et al., 2007), so there is a clear need to evaluate the sensitivity of oligotrophic marine environments to this anthropogenic effect. The Mediterranean Sea is one of the most nutrient-poor waters in the world with maximum open sea area chlorophyll concentrations of 2-3mg m⁻³. Its' trophic status varies from oligotrophic-mesotrophic in the northwestern basin to extremely oligotrophic in the eastern basin (Moutin and Raimbault, 2002; The Mermex Group, 2011). High biological activity occurs annually in parts of the western Mediterranean, including coastal France in the late winter and early spring (D'Ortenzio and Ribera d'Alcalà, 2009; Siokou-Frangou et al., 2010).

In this work, we collected water from three mesocosms deployed in the Northwestern Mediterranean Sea over two campaigns during different seasons as part of the European Mediterranean Sea Acidification in a changing climate (MedSeA; <http://medsea-project.eu>) and the Chemistry-AeRosol Mediterranean Experiment (ChArMEx) projects to test the effects of ocean acidification and changes in the biogeochemistry of the seawater on the physical and chemical properties of primary marine aerosol including size distributions and CCN activity.”

The authors did not emphasize enough what is the added value of pelagic seawater/mesocosm when characterizing primary marine organic aerosol emissions.

Please see the response to the above question.

We have also added the following sentence in Section 2.1 to clarify why these experiments were performed off-shore in the methods section, “For both campaigns, the mesocosms were located off-shore in pelagic waters in order to measure primary marine aerosol properties and biogeochemical parameters of the water while minimizing contamination from anthropogenic sources.”

- A lot of important conclusions (correlations, equations . . .) are drawn based on the use of two datasets (pre-bloom BV and oligotrophic BC) which are very contrasted, showing most of the time two groups of points with respectively high/low values leading obviously to a very good correlation. I am not convinced that it is legitimate to perform such correlation plots unless the authors bring much enough material to demonstrate, for instance, that very similar pre-bloom BV conditions can be observed also at BC and that oligotrophic BC conditions can also observed at BV.

We thank the reviewer for this point. We refer in the manuscript to (Gazeau et al., submitted) currently under evaluation in the journal *Estuarine, Coastal and Shelf Science*, which describes in detail both experiments and the two investigated sites. You can see in the figure below (included in Gazeau et al., submitted) that, indeed, these two bays share many similarities with typical bloom conditions in winter-spring and oligotrophic conditions in the summer corresponding to a stratified water column. Pre-bloom conditions are also observed in BC. We have now added the following to point the reader towards this manuscript, “The two bays share many similarities in term of temperature, salinity, phosphate (PO_4^{3-}), nitrate+nitrite (NO_x) and silicate (Si) seasonal variations, and they both show typical bloom conditions in winter-spring and oligotrophic conditions in the summer, corresponding to a stratified water column. Pre-bloom conditions are also observed at both locations (Gazeau et al., submitted). The presence of pre-bloom and non-bloom conditions was confirmed by the order-of-magnitude difference in the average seawater chl a concentrations ($\text{chl}a_{\text{BC,avg}} = 0.069 \pm 0.009 \text{ mg m}^{-3}$, $\text{chl}a_{\text{BV,avg}} = 1.005 \pm 0.125 \text{ mg m}^{-3}$). Detailed site and experimental information for both campaigns that legitimize the comparison to test in these two locations at different seasons can be found in (Gazeau et al., submitted)”.

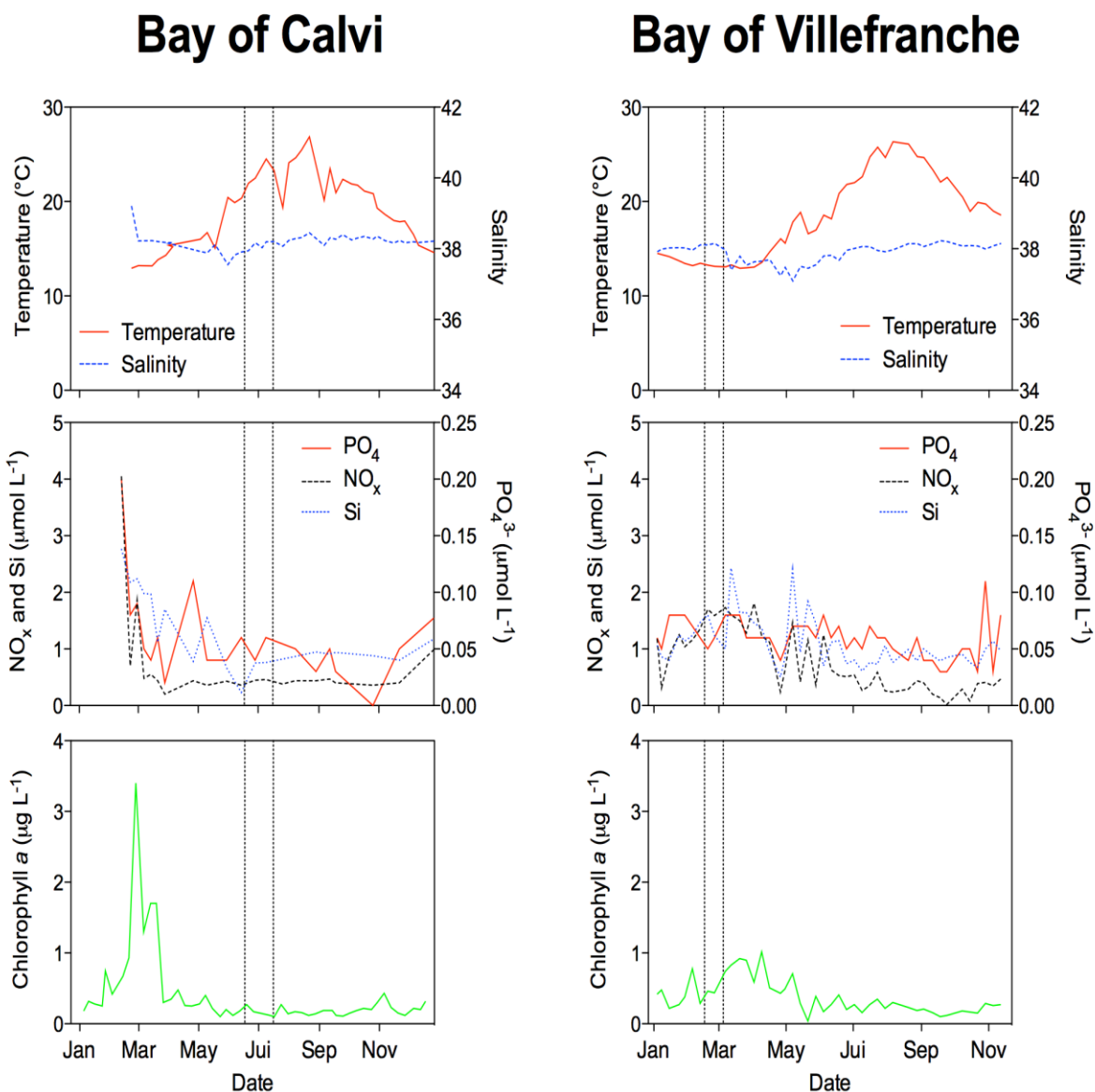


Figure 1. Hydrological (temperature and salinity) and biogeochemical (nutrients: nitrate + nitrate, NO_x ; silicate, Si; and phosphate, PO_4^{-3} and chlorophyll a concentrations) conditions in 2012 in surface waters of the Bay of Calvi (left panel) and in 2013 in surface waters of the Bay of Villefranche (right panel). For all plots, the corresponding experimental periods are identified by vertical dotted lines. [Taken from Gazeau et al., (submitted)].

Minor comments:

Abstract:

Line 43: Be more specific regarding marine aerosol. Uncertainties do concern primary (and possibly secondary) organic aerosol emissions

Acronyms (Dp; pCO₂) to be introduced.

We have made these changes within the text.

Introduction:

Line 75: “. . .via chemical processing”. I would have been more specific (gas-to-particle conversion).

Line 77: “. . .and affecting cloud formation”. Not only. Also cloud properties (albedo, lifetime). I would have added some references here related to the indirect effect of marine aerosols (there are a lot).

Line 82: Should be “primary” marine aerosol. The same in line 103

Line 118: What is “SDS”? Not defined in the manuscript. However, essential to understand some statements.

Line 146: Should be “chl-a” (and acronym explained once in the text)

Line 205: Explain acronyms (MedSeA and ChArMEx)

These changes have been made.

Line 173: Can you explain what you mean by “100x”. 100 times? Can we write in a paper 100x?

The reviewer is correct, “100×” means 100 times the typical ocean concentrations. We used the same terminology that was used within the direct source material (Moore et al., 2011) in order to follow the same convention in describing their results.

Line 174: Are you sure we can investigate in lab experiment cloud formation properties? (not sure).

In a laboratory setting, it is possible to determine whether the tested organics have impacts on CCN activity, through the formation of organic films or decrease in the aerosol surface tension. This can also provide information on water uptake, which is one aspect of cloud formation. The Moore et al. (2011) publication describes their results in terms of CCN activity and cloud formation properties (focused on surface tension depression from surface-active organics and its effect on CCN activity), and we have followed their convention in describing their results.

Materials and methods:

Lines 217 & 223: Be consistent throughout the paper. Seawater or sea water. Not both

Line 226, 246, 259: Explain acronym ETFE, PFA, CTD

These changes have been made.

Line 233: How representative are the concentration of pCO₂? Provide a range in ambient (atmospheric) CO₂ concentrations.

In the Bay of Calvi (BC), the six targeted elevated pCO₂ levels were 550, 650, 750, 850, 1000 and 1250 μatm. In the Bay of Villefranche (BV), the levels were 450, 550, 750, 850, 1000 and 1250 μatm. These pCO₂ levels were chosen in order to cover the range of atmospheric pCO₂ projected for the end of the century following various CO₂ emission scenarios, from the representative concentration pathways (RCP) 2.6 (i.e. radiative forcing of 2.6 W m⁻² with a pCO₂ level of 490 ppm) to the pessimistic RCP8.5 (+ 8.5 W m⁻², pCO₂ ~ 1370 ppm) as seen in the IPCC report (IPCC, 2013). We have clarified this within the text.

Line 312: Can you help the reader and state in one sentence how representative are the SS values you have set in your paper?

We have added the following sentences to clarify this for the reader, “The range of SS values used in this work is typical of those reported in natural clouds. Anttila et al. (2009) found cloud SS values from 0.18 to 0.26% for low-level clouds in Northern Finland, Hegg et al. (2009) obtained a SS range from 0.2 to 0.3% for clouds over the California coast, and Asmi et al. (2012) found SS values from 0.1 to 0.3% at the puy-de-Dôme station in Central France.”

Line 315: Table 1 only concerns BV ? If so, please state it.

Table 1 includes the activation diameters and supersaturation values used for both campaigns, not only BV. We have clarified this in the text.

Line 327: “. . . typically less than 10%”. Provide a reference.

The determination to use total organic carbon measurements and refer to them as dissolved organic carbon measurements was specific for our experiments (given the low concentration of particulate organic carbon). There is no reference, as this is not a generalizable trait. We have added the measurements of particulate organic carbon for clarification.

Line 331: “0.4mm”. Do you mean “0.4mm pore size diameter” ?

We thank the reviewer for this; we have clarified this within the text.

Results and Discussion

Lines 338-346: You just state here that acidification experiments did not show any difference compared to the control, but you do not say if this is something expected or not. This is an important result having strong implication for the paper but it is not commented at all.

We thank the review for pointing this out. It is important to note that a goal of the present paper was to characterize the relationship between seawater biochemical composition and primary marine aerosol properties, independent of the level of acidity. We have added the following sentence to the discussion in Section 3, “While several studies have shown the effects of ocean acidification on biogeochemical parameters in eutrophic waters (Galgani et al., 2014; Schulz et al., 2013), observations from the MedSea experiment showed no effect of ocean acidification on most of the biogeochemical parameters in these oligo- to mesotrophic areas. These results are discussed more fully in Gazeau et al., (submitted). As a consequence, we did not expect any impact on the primary marine aerosol physical aerosol properties.” We have also added more discussion about ocean acidification into the introduction.

We have already included discussion of the acidification experiments later in Section 3.4, “The control and acidified mesocosms showed no significant differences in terms of correlations between organic fraction and different biogeochemical parameters. For studies of marine aerosol, this indicates that any acidification effects on these biological parameters impacts the physical and chemical parameters of the aerosol much less than the natural variances caused by organic pre-bloom and bloom periods. It is not yet clear whether this observation can extend beyond the western Mediterranean Sea.”

Line 352: You give 4 lognormal modes but no information on uncertainties (such as standard deviation). How many data were used / averaged to get these results? These points are clearly missing here.

We thank the reviewer for bringing up this point. We have added standard deviations and additional relevant information to Section 3.1 on the size distribution and lognormal mode fitting. We have also modified Figure 1 to show the envelope in the size distributions for both campaigns.

Line 379. “. . . tested synthetic sea salt with a weir”. Can you clarify. What is the weir?

A weir is another method of forming bubbles to measure the properties of primary marine aerosol. It is fully described in the text of Sellegri et al. (2006).

Lines 348-389: You compare well your results with the literature but you do not conclude. Where is the reality? It would be helpful for the reader to know what is supposed to be representative of sea salt modes. Wave channel experiment is the closest to reality?

In Section 3.1, we have added the following, “Differences in the size distribution of laboratory generated primary marine aerosol found in the literature seem to depend on the method used to

generate them. Fuentes et al. (2010) observed 4 modes (modal sizes 14, 48, 124, 334nm) generated from plunging-water jet experiments with artificial seawater. Mode 4 was believed to be linked to splashing water from the jet mechanism. Plunging-water jet experiments were found to most closely mimic the size distribution of ambient primary marine aerosol (Fuentes et al., 2010) while generating sufficient aerosol for characterization measurements.”

Line 396-397: Again you compare your results with the literature but you do not conclude. Your temperature increases by 2 to 5 times more than the one reported by Zabori et al. So what?

We clarified the following paragraph to showcase where our results were different from previous studies and what this could indicate, “This increase is 2-5× times higher than the 1-2°C hr⁻¹ temperature increase measured by Zábory et al. (2012). Even with this large temperature range, we did not observe that the increase in the water temperature affected the shape of the size distribution. We were not able to make qualitative statements about the relationship between the number concentration and temperature, with the level of uncertainty in the wind flow stability within these experiments.”

Concerning the relationship between temperature and number concentration, we have removed Figure 5 and subsequent discussion from the manuscript. With more analysis, we were not able to make qualitative statements about the relationship between the number concentration and temperature, with the level of uncertainty in the wind flow stability within these experiments.

Line 413: “. . . the enriched samples showed similar behavior to the non-enriched waters”. Hereafter, you compare your results with the literature but again you do not conclude. I would have said that it is somewhat unexpected. No? Please help the reader and provide conclusions.

An explanation of the addition of the enriched sea surface microlayer was already provided within the text in Section 3.2, “Additionally, the enriched samples showed similar behavior to the non-enriched waters, indicating that the addition of the organic-rich microlayer had little effect on the water uptake for the aerosols. For the experiments incorporating the enriched microlayer, the entire organic-rich volume was added a few minutes before starting the water jet system, rather than being continuously introduced to the tank throughout the entire experiment. This could have led to microlayer depletion over the course of an experiment, explaining why no visible difference was seen between the enriched and un-enriched samples. However, no clear difference was seen between the first (when the microlayer was present) and subsequent size distributions during a given microlayer enriched experiment.”

We have added the following statement to the conclusions, “Additionally, experiments including the enriched sea surface microlayer, which increased organic concentrations, showed no marked difference from the un-enriched mesocosm samples, indicating that enrichment did not influence

the water uptake of the primary aerosol at the thermodynamic equilibrium reached in the CCN chamber.”

Lines 429-430: How results at SS=0.39% can be more variable with a standard deviation of 2% compared to SS=0.08% (standard deviation of 7%)?

We thank the reviewer for this comment. We have changed the text to the following, “At BV, the average activation diameter for SS=0.39% over the course of the experiments was $D_{p,50,avg}=59.48\pm 1.1\text{nm}$ (Fig. 5), while at SS=0.08% the average activation diameter was $D_{p,50,avg}=141.91\pm 10.8\text{nm}$ (Fig. 6).

Lines 437-442: I am not convinced by the interpretation of Fig. 6. You are dealing here with very different water conditions showing different organics (chl_a) that is a major factor controlling CCN activity as stated before. The Temperature in Fig. 6 is simply showing the different water conditions (oligotrophic vs pre-bloom). In this Fig 6, we see an anti-correlation at BV (which is more interesting than comparing BC and BV together). However, without a clear view of the temporal variability of other key parameters (Chl_a, . . .) it remains difficult to conclude here that temperature is a driving factor controlling CCN activity.

It is important to note that the temperature of the air shown in Figure 6 is not indicative of the different water conditions (oligotrophic vs. pre-bloom). Bloom periods are dependent upon both the season and weather conditions (multiple sunny days in a row) as well as the nutrient levels in the water and water conditions (water temperature and stratification), but air temperature has not been identified to play a strong factor.

We agree with the reviewer that the idea that air temperature in field experiments could be a factor controlling CCN activity is not well defined and certainly not a conclusion of this work. We have used cautious language to mention this relationship, as it is not previously seen in the literature, and no clear conclusions were drawn from this work. However, we believe it is important to highlight this so that future work may see if in fact the air temperature impacts the emission of small particles. It was outside the scope of this work to study the different biogeochemical parameters dependence on air temperature in detail.

Line 471: Can you confirm that the results from Pringle et al., were also obtained at SS=0.39%? Otherwise, is it legitimate to compare the kappa value at BC with this paper?

Kappa values from Pringle et al. (2010) were calculated as global annual mean values, and were not calculated specifically at one SS value, but instead for a bulk chemical composition. In Section 3.3, we now write:

“In BC, the average mesocosm kappa value at SS=0.39% was $\kappa_{\text{avg,BC}}=0.95\pm 0.17$. In BV, the average mesocosm kappa values for SS=0.39% and 0.08% are $\kappa_{\text{avg,BV}}=0.45\pm 0.13$ and 0.78 ± 0.14 , respectively. This indicates that the smaller particles (measured at the higher SS) were higher in organic material. The kappa values obtained at SS=0.08% are more representative of the kappa that would be obtained for a bulk chemical composition, as most of the aerosol mass is comprised in the accumulation mode for submicron particles. Our value then falls well into the suggested range of the kappa average of marine aerosol, $\kappa_{\text{marine}}=0.72\pm 0.24$ (Pringle et al., 2010).”

Section 3.4: As mentioned above (major comments), I am not convinced at all that it is legitimate to draw correlations from 2 different datasets (BC and BV). It is like drawing correlation plots with 2 points only. Not statistically relevant.

We refer the reviewer to the explanation and figure above. As mentioned there, Gazeau et al., (submitted) contains details and a comparison of the two sites to rationalize comparing the two data sets.

Line 521: “The parameterization derived in this work is a high estimate . . .”. Why ? I cannot find the reason. It is a strong statement since hereafter you propose that chla may not be the only driving force controlling the organic fraction. Later, you mention correlation of $r^2=0.161$ with virus-like. Can we state that r^2 of 0.161 is good enough to depict a correlation? (not sure).

In Figure 7, the parameterization from this work gives a higher organic fraction for a given chla content compared to other published chla-organic fraction parameterizations. This could be due to the size dependence of the organic fraction of the aerosol (with higher organic content for smaller particles) as described in the paper. We now include this in the text: “The parameterization derived in this work lays at higher organic content when compared to other parameterizations, even though it does not include secondary organic contributions as do many of the satellite-receptor site studies. This is likely due to the size dependence of the organic fraction that we observed in the BV data. Our parameterization is applicable for Aitken mode type aerosols and would probably shift towards lower organic content for accumulation mode particles.”

We have also modified our discussion of other driving forces controlling the organic fraction, a hypothesis that is investigated using other biological populations and tracers. We now include, “It is also possible that organic components in Mediterranean primary marine aerosol are of multiple origins and not solely linked linearly to chla-rich species.

We have included all correlations to showcase the complexity of the relationship between primary marine aerosol organic fraction and different biogeochemical parameters. We have now

calculated the significance of the correlation coefficient and added p values for all the correlations. We found that all of the correlations were significant ($p < 0.05$), even when the R^2 values were < 0.5 .

Line 543: Sentence “However, strong anti-correlations were observed between Mode 1 . . .”. I cannot find the figure where we can observe this anti-correlation.

These figures were not included within the paper; we chose only to discuss the correlation between the mode fitting and the different biogeochemical parameters. We have clarified this within the text.

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