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 comments and discussion. The concerns of this reviewer are addressed below.

General comments

Since the main objective of the paper is to compare number size distribution for high and low-productivity episodes, more data/discussion need to be provided for the particle size distribution fittings. This is important part of the paper, yet there is only one figure and one table for " an average size distribution." Based on Fig. 2 I gather that the daily-averaged number fraction of DMPS lognormal modes changed considerably as a function of time for different mesocosms. For example, for BC on 06/29 Mode 1 fraction increased by more than 50% (and modes 2, 3, and 4 decreased correspondingly) for P6. For P3 and C3 changes were not so large. Similar picture is observed for BC on 07/02. The differences were also observed for BV (e.g., 02/23 and 03/03). Since the measurements were carried out continuously, I think the authors should show the errorbars for the variability in the number fraction of DMPS lognormal modes. It is important to clearly demonstrate that this variability in smaller than for example, the differences in Mode 2 particle fractions between BV and BC. Please also show the size distribution fittings separately for BC and BV, accompanied by appropriate statistics. This will help the reader to clearly see the influence of biological productivity on the Aitken mode.

We thank the reviewer for bringing up this point. We have added the size distribution spread for both campaigns in Figure 1, as well as added the mode fitting standard deviation in the text and within Figure 2. We have also adjusted the language at the beginning of Section 3.1 to now read: "The marine aerosol size distributions remained fairly stable during a given experiment, which lasted around one hour for each water sample. The aerosol size distribution also remained stable throughout the course of each campaign, with a similar distribution shape. Four lognormal modes were fit to the average size distributions of each campaign, with results summarized in Fig. 1 and Table 2. The average size distributions were taken from 214 size distributions for BC and 182 size distributions for BV, and size envelopes are also included within Fig. 1. In order to investigate the size of the aerosol independently of the concentration, the size distributions were normalized using the total aerosol number concentration. We found that the primary marine aerosol size distributions were best described using the three expected modes: nucleation mode (around 20nm), Aitken mode (around 40nm), and accumulation mode (around100nm); and an additional fourth mode around 260nm. Using only three modes for the fitting procedure could not satisfactorily represent the primary marine aerosol size distribution. A mode at around 300nm was also found by Sellegri et al. (2006) during bubbling experiments for which the effect of wind on the surface breaking bubbles was simulated in a similar experiment set-up. The

300nm mode was interpreted by the authors as the result of a thicker bubble film where the bubbles are forced to break by the wind instead of reaching a natural breaking thickness. The four average lognormal modal diameters determined (18.5±0.6, 37.5±1.4, 91.5±2.0, 260±3.2nm) were present in both BC and BV."

The discussion regarding the role of organics for the increase in the Mode 2 particle fraction on page 26200, line 26 should either be removed or re-written. Chapter 3.1 gives no information to conclude that the increase in particle fraction can be attributed to organic material. The fact that other studies may have seen similar changes in lognormal mode distributions and attributed that to organics is not enough justification.

We agree that the assumption of organics being responsible for the Aitken particle increase was introduced prematurely in the paper. Indeed, it is the increase of the organic content of the Aitken mode particles reported in Section 3.3 that indicates that the increase in the particle fraction of Mode 2 is likely due to the presence of organics. We have changed to text to better explain this. The text of Section 3.1 now reads:

"Throughout the campaign in BC, the fractions of Modes 1-3 were approximately equal in magnitude (0.297), whereas in BV, the magnitude of the Mode 2 (the Aitken mode) fraction relative to the other modes was dominant (0.48). These same trends were observed for all experiments using SS=0.08% and all the enriched mesocosm samples (Figs. 3-4). When augmenting the bacterial abundance in seawater, Collins et al. (2013) observed an increased particle fraction of the smallest lognormal mode diameter with no change to the shape or magnitude of the size distribution; this was attributed to the replacement of internally mixed salt/organic particle types by insoluble organic type particles. Previous studies have also indicated changing size distributions or mode number fractions with increasing organic material (Fuentes et al., 2010; Sellegri et al., 2006). In the present study, we will examine which chemical component is linked to the increase of the Aitken mode particles in Section 3.3. "

Section 3.3 now includes, "The organic fraction of the Aitken mode particles (obtained from measurements performed at SS=0.39%) is significantly increased during the BV experiment compared to the BC experiment. This indicates that the Mode 2 fraction increase observed in the size distribution is due to the presence of organic matter, in agreement with the observations of Collins et al. (2013)."

I strongly recommend removal of all the discussion regarding ocean acidification. The fact that some CO2 was pumped in seawater tells noting on how the ocean ecosystems, and therefore carbon content/speciation will change in future scenarios of elevated CO2 and changing climate. Such speculations can lead to the erroneous conclusions.

We thank the reviewer for this point, however we disagree. We have expanded the discussion of ocean acidification within the introduction to make it clearer why this study is important and

how it relates to current and past research. 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. Many experiments, with some following the same protocol as in this work (mesocosms deployed for multiple days), have already shown that increased CO_2 availability could lead to important modifications of planktonic community compositions and functioning. Shifts in organic matter production (increased dissolved form concentrations with increasing CO_2) will potentially affect marine aerosol production. We agree that our experimental protocol does not fully reflect the environmental conditions of the ocean in 100 years, mostly because we do not consider other climate change related drivers such as temperatures changes, precipitation levels, changing wind speeds, and more. However, we do believe that performing such experimental work as described here has the potential to bring invaluable information that would ultimately need to be incorporated into models in order to refine our predictions for the future decades.

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."

I do not believe I understand Figure 5. How were the error bars calculated? If there are no vertical error-bars (like for many P6 measurements) does that mean that within a day Ntot did not change at all?

We thank the reviewer for bringing this to our attention. We have decided to remove 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.

According to the methods description, the covers were elevated to ~10 cm above the top of the mesocosms, allowing air to circulate to avoid a confinement effect in the trapped water. I am just curios what was happening during "dangerous wind and wave conditions?" Unless some additional precautions were taken wouldn't seawater spillover into the mesocosms?

"Dangerous wind and wave conditions" occurred when sampling would have put researchers on rough seas with strong winds. The reviewer is exactly right about spillover, and in fact, sampling was halted for the BV campaign due to seawater spillover into the mesocosms from the high wave conditions. However, for the data shown here for BC and BV, based on the consistency of CTD measurements, there was no observed encroachment of seawater into the mesocosms.

Specific comments

Page 26190, Line 4 (and elsewhere): Please change "sea salt" to sea spray when referring to primary marine aerosol emission.
Page 26190, Line 6: Please note that you are referring to particle diameter when using Dp.
Page 26190, Line 22: Please use the plural form of "organic".
Page 26193, Line 17: Please change "lower diameters" to "smaller diameters".
Page 26198, Line 24: Please define the two supersaturations used here and provide the reason behind the selection.
Page 26198, Line 24: Please add the temperature scale to "6◦". Same for "3◦" in Line 27.

These changes have been made.

Page 26199, Line 17: Please provide the data for particulate organic carbon concentrations to justify the argument that measured TOC is reported as DOC.

We have added this information to the text. The text now reads, "Total organic carbon (TOC) was measured instead of dissolved organic carbon (DOC) in order to avoid contamination during filtration. However, the TOC measurement is referred to hereafter as DOC, due to the low concentration of particulate organic carbon in both sites (averaged over all mesocosms and all sampling times, BC: 4.32 ± 0.91 μMol, BV: 11.49 ± 5.50 μMol, which was typically less than 10% of TOC)."

References:

Collins, D. B., Ault, A. P., Moffet, R. C., Ruppel, M. J., Cuadra-Rodriguez, L. A., Guasco, T. L., Corrigan, C. E., Pedler, B. E., Azam, F., Aluwihare, L. I., Bertram, T. H., Roberts, G. C., Grassian, V. H., and Prather, K. A.: Impact of marine biogeochemistry on the chemical mixing state and cloud forming ability of nascent sea spray aerosol, *Journal of Geophysical Research: Atmospheres*, *118* (15), 8553-8565, 2013.

Fuentes, E., Coe, H., Green, D., de Leeuw, G., and McFiggans, G.: On the impacts of phytoplankton-derived organic matter on the properties of the primary marine aerosol – Part 1: Source fluxes, *Atmospheric Chemistry and Physics*, *10* (19), 9295-9317, 2010.

Sellegri, K., O'Dowd, C. D., Yoon, Y. J., Jennings, S. G., and de Leeuw, G.: Surfactants and submicron sea spray generation, *Journal of Geophysical Research*, *111* (D22), D22215, 2006.