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.

F. Dulac

We kindly thank Mr. Dulac for his insightful comments. We address particular concerns below.

- My most important concern is about particle size distribution fitting and related results discussion. Even if those numbers are stated as approximate by using the sign "~", I cannot find realistic to report with 3 significant digits either mode fractions (e.g. ~0.295, 0.482) or mode diameters (~18.5, 37.5, 91.5 and 260 nm). Table 2 suggests for instance that it would probably be more appropriate to give ranges for every mode. In addition, we miss information on the variability on the geometric standard deviations of the fitted modes when they are needed to fully characterize the size distribution. I believe that we miss in the methodological section 2 a sub-section describing the particle size distribution fitting technique and discussing accuracy of results which are the basis of the paper. This is important that the limits of this fitting have been assessed when discussing differences between experiments. I would also expect to see discussed how robust is the choice of 4 modes for fitting, and how variable are the size distribution results. For instance we miss associated standard deviations in Table 2 and I find that Figure 1 should be completed with another plot that shows the variability (e.g. by plotting all curves obtained on a single plot, or showing envelopes...).

We thank Mr. Dulac for bringing up these points. We have decreased the number of significant digits and included the standard deviations for the mode diameters. We have added standard deviations for the mode fittings in Table 2 as well as the size distribution spreads in Figure 1.

We have also changed the first paragraph in Section 3.1 to describe the size distributions and mode fitting in more detail. It now reads, "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."

- In section 2.2, I find that we miss information of DMA size channels and measurement integration time for size distributions. How variable is the size distribution measured for a set of given conditions?

We have added information about the number of channels for the DMA measurements and the size stepping integration time and the stability of the size distribution over the course of an experiment. We have also added information about the dataset size for the average size distribution measurements.

- I am questioning the negative values of kappa occasionally found. Cannot you tentatively use them to better constrain Kappa-inorg? If you really end up with unrealistic values for Kappa-inorg, is it justified to keep those points in the data set for correlations rather than discarding them (figures 9-10)?

The Kappa-inorg value is already set to the theoretical kappa value of inorganic sea salt. The variability of the kappa-total measurements around this theoretical value creates unrealistic negative organic fractions. These negative fractions give us the level of uncertainty in the method to determine organic fractions and we believe that they should be kept in the data set.

- When discussing correlations with biological parameters in section 3.4, I find that you might be more precise by giving significance levels of the correlations (that depend on the number of points used or degree of freedom). Is it really appropriate to state that you "find a correlation" when R is about 0.4 or a bit lower. In the case of the correlation between Synecococcus and DOC, is it really robust or rather driven by the single point with the highest DOC value: what if you fit without this peculiar point? The sigmoid fit used in Fig. 10f seems also more appropriate for most of the correlations (e.g. Fig. 10a, 10c, 10e): is it justified to stick to linear correlations?

We thank Mr. Dulac for bringing up this point. We have 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. We also believe that the sigmoid fits are more appropriate, especially conceptually because the organic fraction of the PMA cannot be higher than one even after reaching a certain level of chla content. We now mention this in the text, and we have included sigmoid fits for all the correlations, where applicable. However, in order to compare our results with results reported in the literature, we also provide the linear correlation fits.

The following paragraph is now added to Section 3.4: "Sigmoid fits are also shown in Figs. 8 and 9 for all biological parameters where they could be determined. Sigmoid fits might be more appropriate to use in many cases, to conceptually constrain the organic fraction of the primary marine aerosol to one regardless of the chla concentration. We have included both linear and sigmoid fits, with their respective R^2 and χ^2 values for completeness."

Other technical comments:
-You should generalize the use of the italic style for all symbols throughout the paper.
-Abstract: it would be worth indicating the particle size range of your measurements (10-400 nm) which constrains the size distribution fitting.

-Abstract, line 16: specify "kappa (k)".

We have made these changes.

- p.26196, line 16: given that the mesocosm diameter is 2.3 m, I rather calculate that 5 L of water corresponds to 1.2 mm in height, not 15 cm.

We apologize for the confusion. The 15cm indicated the maximum depth to which we collected the surface water. We have clarified this within the text.

-p.26197, l.11: I think "cm^3" is expected instead of "cm"; I would rather write "of ~10 cm".
-p.26202, l.6: specify "the water temperature".
-p.26203, l.12: I'd rather use the singular in "no visible changes".
-p.26206, l.12: specify that this correlation is "(not shown)".
-p.26207, l.25: I'd rather use the singular in "any parameters".
-p.26208, l.25-26: the second part of the sentence seems unclear to me as written; I think that the two word groups "Chl a and additional pigments" and "the organic fraction" must be switched in the first part of the sentence.
-Acknowledgements: the acronym MISTRALS is missing its terminal S; "MISTRALS/ChArMEx" would be more appropriate.
-Fig. 1: Ordinate axis legend should probably state "Normalized number fraction", since the curves appeared normalized by their maximum value.

These changes have been made.

-Table 2: It is not clear in the legend that these numbers are averages.

We thank Mr. Dulac for this point. We have clarified the text to indicate that these are averages.

-Many figures are difficult to read once printed, with too small characters.

We have increased the fonts of all figures to make them easier to read.

-Legend of Table 2, Figures 3 and 6 (and possibly other occurrences): I would specify "CO2-enriched".

We apologize for the confusion. "Enriched" in all of these contexts indicate that the samples were enriched with the sea surface microlayer. We have tried to clarify this within the text where necessary.

References:

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.