Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-320-AC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Primary marine aerosol physical and chemical emissions during a nutriment enrichment experiment in mesocosms in the Mediterranean Sea" *by* Allison N. Schwier et al.

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Received and published: 7 September 2017

The authors would like to thank both reviewers for careful review of our manuscript and providing us with their comments and suggestion to improve its quality. The following point –by-point responses have been prepared to address all of the reviewers' comments.

Reviewer 1: Abstract. I would number the main conclusion of the paper, the abstract now is too general and I would state what the main results are such as (1) SMPS (2)

CCN (3) chemistry (4) correlations (poor) with biology.

Answer 1: the abstract was re-written as suggested by both reviewers. It now reads:"While primary marine aerosol (PMA) is an important part of global aerosol total emissions, its chemical composition and physical flux as a function of the biogeochemical properties of the seawater still remain highly uncharacterized due to the multiplicity of physical, chemical and biological parameters that are involved in the emission process. Here, 2 nutrient enriched-and 1 control mesocosms filled with Mediterranean seawater were studied over a three-week period. PMA generated from the mesocosm waters were characterized in term of chemical composition, size distribution and size segregated cloud condensation nuclei (CCN), as a function of the seawater chlorophyll-a (Chl-a) concentration, pigment composition, virus and bacteria abundances. The aerosol number size distribution flux was primarily affected by the seawater temperature, and did not vary significantly from one mesocosm to the other. The aerosol number size distribution flux was primarily affected by the seawater temperature and did not vary significantly from one mesocosm to the other. Particle number and CCN aerosol fluxes increase by a factor two when the temperature increases from 22 °C to 32 °C, for all particle submicron sizes. This effect, rarely observed in the literature, could be specific to oligotrophic waters and/or to this temperature range. In all mesocosms (enriched and control mesocosms), we detected an enrichment of calcium (+500%) and a deficit in chloride (-36%) in the submicron PMA mass compared to the literature inorganic composition of the seawater. There are indications that these chloride deficit and calcium enrichment are linked to biological processes, as they are found to be stronger in the enriched mesocosms. This implies a non-linear transfer function between the seawater composition and PMA composition, with complex processes taking place at the interface during the bubble bursting. We found that the artificial phytoplankton bloom did not affect the CCN activation diameter (Dp,50, average=59.85$\pm$3.52nm and Dp,50,average=93.42$\pm$5.14nm for sursaturations of 0.30% and 0.15% respectively) nor the organic fraction of the submicron PMA (average organic to total mass= 0.31$\pm$0.07) compared to the control mesocosm. Contrary to previous observations

in natural bloom mesocosm experiments, the correlation between the particle organic fraction and the seawater Chl-a was poor, indicating that Chl-a is likely not a straightforward proxy for predicting at the daily scale PMA organic fraction in models for all types of sea and ocean waters. Instead, the organic fraction of the Aitken mode particles were more significantly linked to heterotrophic flagellates, viruses, and dissolved organic carbon (DOC). We stress that different conclusions may be obtained in natural (non-enriched) or non-oligotrophic systems. "

Reviewer 2: I would stress the correlation with biology (Figure 8) were poor. - I would add a paragraph on the discussion on the time resolution of the filters used (which could not find it on the paper) and if these types of measurements can be useful, or if we need to move to new types of instruments (on line such as ATOFMS, HR AMS, etc) to actually see any valuable variation over time.

Answer 2: The time resolution of filter measurements is one hour, the same that time resolution of each single bubbling experiment. On-line chemical measurements would require that aerosol is continuously generated with a continuous seawater flow in the bubbling device. This would be of high interest to observe diurnal variations of processes occurring. The conclusions of this paper rather points towards a need for longer time periods of observation rather than to a larger time resolution though.

Reviewer 3: I would also stress over the text more the importance of the regionality (point well made) where the Mediterranean oligotrophic waters need more attention, and other mode studied marine aerosol areas may be different.

Answer 3: We did stress this aspect more clearly throughout the paper, mainly in the discussion part.

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