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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.

Anonymous Referee #1

Received and published: 31 December 2014

This study reports important information on primary marine aerosol emission properties (organic fraction, size spectra, CCN activity) for two mesocosm studies performed in the Mediterranean Sea during two contrasted periods (pre-bloom, oligotrophic). This study also presents results from acidification that poorly affect primary marine organic aerosol emissions. A new equation is proposed here to describe the fraction of organic in marine aerosols as a function of chl a . This paper is well written and results are properly compared with literature data. A nice overview of the factors controlling primary organic aerosol emissions is provided in the Introduction. However, results obtained

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here are, for some of them, not convincing. I have few major concerns that need to be addressed properly before considering this paper to be published.

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

- 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.

Minor comments:

Abstract:

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

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Acronyms (Dp; pCO₂) to be introduced.

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

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

Line 205: Explain acronyms (MedSeA and ChArMEx)

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

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

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

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

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

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

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.

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.

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

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 ?

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.

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%)?

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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.

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 ? 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.

Line 521: “The parametrization derived in this work is a high estimate ...”. Why ? I cannot find the reason. It is a strong statement since hereafter you propose that chl_a 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).

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

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 26187, 2014.

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