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Atmospheric
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Discussions

Interactive comment on "Artificial primary marine aerosol production: a laboratory study with varying water temperature, salinity and succinic acid concentration" *by* J. Zábori et al.

Anonymous Referee #1

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The paper aims at addressing important aspects of primary sea spray production like temperature, salinity and effects of organic matter all of which can alter the production mechanism substantially. Conceptually and technically the paper is important, however, apart from the temperature dependence other aspects of primary production are poorly addressed. The paper can not be recommended for publication in ACP in its current form until the major critical points are adequately addressed by revising the paper substantially.

The paper suffers from several key weaknesses.

MQ water has no relevance within the scope of the paper, especially that it wasn't ultra

pure given the number of generated particles. Any natural fresh water would have been a better choice. I would suggest removing most of MQ water data, especially from the graphs where they crowd the plots and scatter focus. MQ data can be moved to supplementary material if authors wish so.

Choosing NaCl for the experiments instead of sea salt or real oceanic water was a bad choice. A dilution of the real oceanic sea water with MQ water for studying salinity effect would have been a much better choice having a direct connection with the freshening of the Artic Ocean water due to Greenland ice sheet melting. Authors put relevance of their results to the future changes of the Arctic environment (increasing temperature and decreasing salinity), however, the set-up of the experiment focused on unrealistic salinity changes. What is the chance of salinity dropping even below 30 g/l with a full melt of the Greenland ice sheet? What type of water body would salinity of 18 g/l represent? It would be extremely important to demonstrate if there were any significant or discernable changes in size distribution in the salinity range of 30 to 35 g/l. The absence of the difference would be as important as the presence of it. Moreover, the data between 18 and 35 g/l can not be linearly parameterised for modelling purposes. Why only 3D distributions are presented in Fig.7 instead of focusing on quantitative parameters between different salinity ranges, again emphasizing the range of 30-35 g/l.

Selection of a succinic acid and particularly its concentration range are completely unrealistic. The succinic acid is indeed ubiquitous in aerosol phase (among many other dicarboxylic acids), but is present in the ocean at concentrations of just ~0.1uM (1ug/l) as authors reference it themselves. Either justified candidate chemical compounds should be used for the experiment in the laboratory or the chemical soup produced by a biological system (e.g. phytoplankton culture) as in Fuentes et al. (2010a,b) should be used. The use of artificial compounds like SDS, Triton-100, etc. is a thing of the past and has little value for the real world system. Unrealistic succinic acid concentration can be equalled to artificial compound. The real ocean is much more complicated anyway than the artificially simulated system in the “jar”. Global range of DOM is in the range of 50-150uM with few exceptions. The effect of 2446uM concentration on the aerosol production has no scientific value as it is completely unrealistic and should be substituted in the abstract with e.g. the rate change – particle number per 10uM concentration change or only the effect of the lowest (realistic) concentration of 94uM is presented. The above problem is even more acute due to the fact that the data with max organic concentration is presented in the graphs which have no connection when compared to the real Arctic Ocean water. Presenting 94uM concentration data can at least be justified on the basis of being of similar molar concentration range as in the open ocean.

Presentation of the graphical information is inconsistent and often out of focus. The focus should be on the size distribution, not the total concentration. The total concentration dependence can be presented as a summary, but actual size distributions are more important for modelling purposes and for comparison with other similar studies from different oceans in the future. The number of nanometer size particles (10-20nm) can differ greatly depending on solution concentration (salinity) (refer to Fig. 7 at 12-18 g/l) while primary sea spray particles in the 10 to 20nm range are very rare in the real oceanic environment. It is important to demonstrate that CPC 3010 agrees well with the DMPS before discussing the graphs referring to authors statement that number of particles in the size range above 0.25um was negligible and therefore, DMPS and CPC must agree well. Fig.7 is only useful as supplementary material as it provides little quantitative information on the observed differences. Moreover, it does not emphasize realistic data. 3D figures are useful for examining evolution not process analysis where small details matter most.

Specific comments

Tables 1-5 can be incorporated into the text as a technical information. Additional Tables are recommended documenting size distribution parameters of most relevant salinities and succinic acid concentrations. I would also recommend fitting the most

relevant size distributions with log-normal modes and presenting modal parameters in the Table. Fitting can only be done after resolving Fig. 6 problems below.

Fig. 3 Why the data of Arctic ocean water are scattered enormously producing wildly different total concentrations at neighboring temperatures? Why absolute concentrations of Arctic Ocean water can not be compared directly with NaCl solution of 35 g/l representing real world ocean salinity? Fig.3 looks inconsistent and the above issues need to be addressed.

Fig.5 can be omitted with ratios summarised in the text as this information is rather arbitrary (depends on the selected size ranges) and of secondary importance.

Fig. 6 is inconsistent with many literature data (e.g. Martensson et al., 2003; Sellegri et al. 2006; Tyree et al., 2007, Keene et al., 2007; Fuentes et al. 2010a and numerous more) where the main sea spray mode is at around 100nm and not 200 nm and above as authors present it. I suspect that particles were not sufficiently dried (only at around the efflorescence point) producing dubious jump around ∼200nm. This can be due to the humidity/sizing effects in a non-insulated DMA column acting partially as CCN chamber in the presence of water vapour (not sufficiently dried air and particles). I also understand that OPC data were corrected/merged with DMPS data in the overlapping range. How the data would look like when OPC data is plotted as it were recorded? If not that jump, the main size mode would have been at 100nm. At the moment higher diameter mode obtained in this study is speculatively attributed to a jet system which was never demonstrated to produce a dominant mode above 200nm (Sellegri et al., 2006, Fuentes et al., 2010a) unless forced breakdown of the foam as in Sellegri et al. 2006. If surfactants were to be responsible for the dominance of >200nm mode, why ALL size distributions in this study have dominant >200nm mode even the ones representing the purest of NaCl solution? I suspect the same problem was present in Hultin et al. 2010 paper as I suspect the same experimental set-up was used. This is a very serious issue which needs to be thoroughly addressed before reconsidering the paper. The experimental set-up should be repeated with a fully dried (∼10%RH) sea salt aerosol. Authors can consult HTDMA system users where fully dried aerosol is at the heart of the method.

Page 19045 line 22. Why stirring was needed to maintain homogeneous salinity once it is established?

Page 19048 line 14. The reasoning of the umbiguity of optical spectrometer data requires more detailed discussion and supporting evidence.

I have many minor comments which are irrelevant given the fact that major changes are required to improve the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 19039, 2012.