Review of “Effects of temperature and salinity on sea-spray-aerosol formation simulated with a bubble-generating chamber” by Sofieva et al.

This manuscript presents a new tank for measuring bubble-mediated aerosol formation from a single layer of bubbles at a water surface. To my knowledge, this is the first tank of this type, and it would be nice if the authors could include a short literature review explaining how the tank is similar/different to other setups.

The authors present experiments where aerosols are generated from artificial seawater with varying salinities at different temperatures and using different bubble flow rates. The topic is timely as sea spray aerosol is a major source of atmospheric aerosols with impact on global climate and the dependence of sea spray aerosol flux on sea water temperature and salinity is not well constrained.

I thus find the topic of the manuscript interesting and timely and welcome a new setup for this purpose. Having said that, I also find the manuscript lacking information or discussion in several places. The authors say several times that they provide theoretical expectations and that they compare with other studies. As outlined below I find that the authors more provide an interpretation of their data, and I do not really see, for example in figures that the authors compare with other studies. This might also be very difficult due to the different mechanisms that may dominate depending on how bubbles are generated.
I thus have a number of concerns and comments, which I find should be addressed before the manuscript can be published.

Overall:

Figures – I find that the quality of the figures should be improved. The figure captions should include more information – which experiments, assumptions etc. font sizes, legends should be made similar across figures.

Text: I find the manuscript somewhat difficult to follow in several places and that detailed and quantitative information is missing. I provide examples below and some suggestions for how to improve.

Abstract:

I recommend rewriting the abstract to provide more specific information. For example, it is not clear what is meant by “previous experiments” mentioned more than once in the abstract.

Introduction:

Please specify what is meant by “large” and small droplets in terms of diameter ranges.

There is a long discussion in the introduction about film and jet drops – it would be relevant to relate to the current study, what kind of drops do the authors form or expect to form?

“When SSA is generated in laboratory conditions, the challenge is to mimic the key processes of the real environment: bubble bursting and initial aerosol generation” I do not understand the meaning of this sentence – is bubble bursting not the initial step in bubble mediated aerosol formation?

Line 109: the addition of surfactants to a sea spray tank and the effect on CCN properties was also elucidated in other studies (e.g. 1-3)
I suggest to change the wording in line 123: “The aim of the current study is to re-evaluate the SSA production as a function of water parameters and verify the findings with basic analytical considerations.” The authors present a new experimental setup and new (and interesting) data from it – these may complement existing data from other labs, but I do not really see that the authors re-evaluate something. Also I do not see that the authors “verify” the data, or at least it should be explained what is meant. As I can see, the authors interpret data. Later it says “results of experiments are compared with theoretical expectations”. It is not clear to me what the theoretical expectations are.

Line 127: what are the “most-important parameterizations” - please provide references.

2.1: The lid is not shown in Figure 1.B

Line 135: large compared to what? It would be useful to provide some references.

I have a few technical questions: What type of glass is the chamber made of? Line 142: what is the length of the capillary – or what is the distance from the tip to the water surface?

What are the “selected capillary parameters”? Line 145: “compared to the sinter filters” which filters are the authors thinking about here?

Line 145: did the authors also test sintered filters – or is this a general comment based on literature? If so, references should be added.

Line 153 – what is meant by appropriate pressure balance? Is it atmospheric pressure inside the chamber or is it different?

Flow rates to the instruments should be given.

Regarding the OPC – why did the authors not apply the shape correction of 1.1 to the data shown in the figures?

How long time were the sea water and SML samples stored frozen before the experiments were performed? At what temperature were they kept?
Please state the supplier and purity of the NaCl used.

Fig C1: I suggest the authors also show photos from the highest flow rate used to demonstrate that still only a single layer of bubbles is formed and also the area of bubbles for comparison to the 0.2 l/min case.

Figure B1: please indicate the sampling flow rates of the instruments. And the ranges for the bubble generating air flow and the supplementary air added.

Regarding the images – five images were chosen for each experiment – why this number? - what was the time between the photos - were there variations in bubble number and foam area over the time of an experiment (30-60 min)?

It is stated that the upward particle flux in the chamber was not higher than 0.2 m/s. How was that measured /calculated?

Line 215: the authors compare with a dry deposition rate "The flux speed less than 1 mm sec-1 is lower than dry deposition velocity of any of the produced particles (Kouznetsov and Sofiev, 2012)" – please explain this a bit more detailed – do the authors assume deposition onto a smooth water surface or how is this obtained?

With a bubble flow rate of 0.01 L/min the dilution flow much be very high for all the instruments to get enough air? Please state the dilution flow and the flow rates to the instruments.

Table 2: I suggest a column with the dilution ratio – how much air was added to the headspace in each type of experiment. This could also be included in the column Varying parameter – when the bubble flow rate is low the make up air must be correspondingly high?. I assume this dilution accounted for in all figures shown (number concentrations)?

Line 220: it would useful also to provide the salt concentrations in g/L in all places.

Line 229: “but still limited single-bubble-thick foam area” – how large a fraction of the surface is covered in bubbles under “limited” conditions?

Regarding the temperature control. It says that the temperature rose 1-2°C during each
measurement. I think thus in the legends it would be more appropriate to give the temperature range for each measurement, i.e. 2.5-4.5 °C, 5-7°C, etc.

Results:

It is nice that the authors provide a list of notation. I still have some questions to the notation however:

Page 12: what is “characteristic” bubble size, please define.

Db is bubble diameter at the breakout plane – this should be clear also from Appendix A.

Dcap = 1.2 mm is the inner diameter of the output nozzle.

How is bubble foam lifetime defined?

“These parameters are important for the follow-up construction of a physical model of the sea spray generation. Wherever possible, the experiments are presented together with basic theoretical considerations highlighting the controlling mechanisms and suggesting the shapes of the key dependencies” I do not see that the authors have constructed a general physical model of sea spray generation – or is this perhaps specifically for this setup?. I would thus suggest to rephrase this statement.

269: The authors write: “if kinematic effects of the outgoing airjet can be neglected” – it would be nice with a bit of an explanation for this statement. Also it is later in the section discussed that kinematic effects are important at higher flow rates. Here it should be stated for which flow rates (given explicitly with values in L/min) kinematic effects are important and for which not.

I suggest to show the breakup plane in the figure, and the forces acting to explain equation (1) – or alternatively provide a reference to a textbook or other where equation 1 is derived. Is the pi in equation (1) related to an angle relative to the break-out plane?

The authors discuss both an upward facing capillary (shown in fig 1B) as well as other directions – but only the upward facing is used as far as I understand and I suggest to remove discussion of the downward/sideways capillary (e.g. line 291-302).
I am a bit confused – in the section lines 307-315 – the authors say that they use equation 2 which is for non-upward facing capillary and refer to figure 1. At the same time they have stated that all results shown are for upward facing capillary.

Line 287: As I understand the volume obtained from equation 1 is the volume of the bubble immediately after it has detached from the capillary. At this point the bubble may not be spherical. The author write that the bubble gets spherical and changes its volume on the way up – how can it change its volume? The authors give a fixed value of 28 mm³ – how is that value obtained and how can it be constant across air velocity ranges?

Similarly - how is the theoretical value of 3.7 mm obtained ? and how is the experimental mean size of. 3.74 mm obtained - I assume it is from the images?

Line 290: what is “artificial water”?

Figure 1: Please explain what is shown in the figure. What do the lines show (Q1 and Q3) and is it the median shown also? Are the end points min and max values? If so, why are there data points higher and lower than these?

Line 312: what exactly is meant by characteristic bubble size ? is it from the photo’s? or from calculations?

Lline 322: what do the authors mean by “aged” bubbles?

Figure 2: please write more information in the figure caption. What was in the tank ? water, temperature. It says that a more precise fitting was made for all salinities – why are not all the data shown in the figure along with the precise fit?

What is meant with “saturation” - does it mean the foam fills the whole surface?

How do the two parameters describe the geometry of the experiment – what is their physical meaning?

Line 332 It would be easier for the reader if the wording was more consistent – is flow
scale the same as the bubble air flow rate?

Please explain the symbols. I assume $A$ is the bubble area and $F$ is the flow rate.

But then for a flow rate of $F=0$ the bubble area is equal to 0.72 cm$^2$ – and it should be zero?

Also – line 340 – the bubble diameter is assumed to be 7 mm - but it has just been shown that it’s not constant with flow rate?

Figure 4: please explain what the “instability range” of the system is.

Line 360-365: “Typical size distributions obtained in the experiments have two distinct ranges: $D_p$ smaller and larger ~2 mm (Fig. 4). These ranges roughly correspond to the different mechanisms of the particle formation: film and jet-originated bubbles (Monahan et al., 1986).”

There is more recent work showing that indeed jet drops do also produce a significant amount of sub-micron sized droplets. This would perhaps be interesting for the discussion.

It should be stated how the authors see the two size regimes – I assume it is the decreasing number with size above 2 micrometer? Please explain/discuss. Why would such a trend relate to film and jet drops?

Regarding production of particles from MQ water – it is known that atomizing MQ or otherwise purified water forms particles when atomized. LaFranchi et al. provided one possible explanation. Perhaps it is relevant to note.

Regarding the data on salinity it would be relevant to compare with the recent work of Zinke et al.

Figure 6: perhaps it is the quality of the figure – but I do not see any triangles (APS) in
I suggest the authors expand a little on what can be concluded from Figure 6. For a given salinity the formation of particles seem to increase with decreasing temperature and more so for the smallest and largest particles? This could be discussed in relation to observations already reported in the literature. It is broadly the same trend for the two salinities?

Section 4.1: I find this section somewhat speculative in several places with quite bold statements: “Supporting the representativeness of the above results, the experiments of Fuentes et al. (2010), and Sellegri et al. (2006) confirmed that a bubbling tank with a water jet system can closely mimic the actual oceanic distribution of the emitted bubbles and aerosols.” and “Therefore, confirmed replicability of our main conclusions from the artificial NaCl solution experiments to Mediterranean and Baltic Sea water is significant.”

Regarding the discussion on bubble lifetimes – the lifetime may depend on many factors and be very different in pure water and salt water. It would be interesting if the authors could comment on that.

Figure 9: what are the uncertainties in the bubble lifetimes? I suggest to show these on the plot.

Line 533-536: I think the text should be rephrased for clarity – two droplets are not identical if they have different salt content? Also do the authors mean “dry” diameter rather than “effective”? Would density of the sea water also be a parameter to consider?

In general, when comparing with literature it should be made clear which studies have used artificial seasalt and which NaCl. Also the bubble generation method: single bubble studies, bubbles generated with a frit/diffuser and with a jet may give very different results due to the generation method. This work represents bursting of a single layer of coagulated bubbles as I understand. It is touched upon in 4.5 but in general some thoughts on how bubble lifetimes, size and particle production are expected to be comparable or different with the method used in this study from other methods would be relevant and interesting.


