Response to anonymous Referee#1: Zábori et al., 2012.

The authors thank anonymous referee#1 for insightful comments on the manuscript. The reviewer provided several suggestions for improving the readability and quality of the manuscript. We have followed the suggestions, and our detailed response is outlined below.

1. Comment:
The seawater properties which the authors want to investigate are water temperature, salinity, and organic content (lines 12-14, p. 16089). Oxygen saturation is chosen as a marker for the organic content (line 27, p. 16089), but no connection between the two is established. Need to add couple of sentences and references showing that oxygen saturation is a reasonable proxy for the organic content in the seawater. Should mention that there are other such proxies, and why the authors have chosen oxygen saturation versus the others.

1. Reply:
We did not assume the oxygen saturation in the sea water as a proxy for the organic content, but the way our text was formulated could give this impression (lines 12-14, p 16089; line 27, p 16089), and it has therefore been rephrased. Thank you for drawing our attention to that.
In lines 24-25, p 16089 we write that "Changing environmental conditions will influence the fauna and flora in the Arctic Ocean […]". This will influence the photosynthesis and respiration and therefore change the oxygen concentration and oxygen saturations in the water column (Boyer et al., 1999; Kester & Pytkowicz, 1968; Falkner et al., 2005). In a similar experiment to our study, Hultin et al. (2011) showed how the dissolved oxygen in the water followed a diurnal cycle driven by the photosynthesis, and how the sea spray production was influenced by this. A reasonable consequence of a changing photosynthesis and respiration is an increase or a decrease of the carbon production, which can lead to a change in the organic content in the water. We consider the dissolved oxygen as a marker of the photosynthesis/respiration, but do not regard it a good measure of the organic content. The actual relation is most likely to complex for a successful direct correlation. In our manuscript we are unable to examine a direct connection between the organic content and sea spray aerosol properties, because we lacked such instrumentation.
We are aware that the oxygen saturation is not only a consequence of respiration/photosynthesis, but is as well a function of water temperature and salinity, the degree of ventilation with the atmosphere by wind and waves, and mixing with differently saturated oxygen water bodies before the sampling took place (Boyer et al., 1999; Kester & Pytkowicz, 1968; Falkner et al., 2005). Nevertheless, what influenced the actual oxygen saturation in our experiment is not critical to our study. We wanted to examine if we can expect a change in sea spray production with a change in oxygen saturation in the water and we wanted to show if this impacts on particle properties. Unfortunately, the oxygen saturation did not change enough during our experiment to be able to examine the influence of the oxygen saturation on the bubble properties (lines 20-24, p 16095).

1. Revision:
In the manuscript it was written:

”Changing environmental conditions will impact the fauna and flora in the Arctic Ocean and thereby also the organic content (Wassmann and Reigstad, 2011; Tremblay et al., 2011).” (lines 24-26, p 16089)
It was changed and augmented to:

”**This study focuses on physical changes of Arctic Ocean water and their impact on aerosol production. We recognize that** changing environmental conditions will impact the fauna and flora in the Arctic Ocean (Wassmann and Reigstad, 2011; Tremblay et al., 2011) **which will have both physical and chemical impacts on the Arctic Ocean water properties. As the flora changes, biological activity may be altered and therefore it is likely that changes in photosynthesis and respiration will occur as well. This photosynthesis/respiration change will impact on the oxygen saturation in the water, since the production of oxygen in the ocean by photosynthesis or a consumption of oxygen by respiration is given (Boyer et al., 1999; Kester & Pytkowicz, 1968; Falkner et al., 2005). The experiments by Hultin et al. (2011) suggested that diurnal changes in dissolved oxygen, caused by photosynthesis and respiration, modulated the sea spray formation. In addition, changes in the chemical composition of the water may arise as changes in photosynthesis and respiration alter the carbon content of the water. The additional consideration of changes in the chemistry of the water goes beyond the scope of this article, but this should be an important question for future studies.**”

2. Comment:
This is a study of SSA production from Arctic seawater samples collected in wintertime conditions. The importance of sampling Arctic seawater is understandable; the Introduction gives a good account of the Arctic amplification in terms of the possible feedbacks that cause it. But the emphasis on wintertime sampling (the title and line 3, p. 16090) versus samples collected at any other time of the year is not clear. One can ask why not sampling Arctic seawater during summer when the maximum warming occurs (lines 29, p. 16088) and represent the future conditions for which the authors want to get implications? If the reason behind the sampling in wintertime is the seasonal delay of the impact of the summer warming on the polar climate (due to the large heat capacity of the ocean, e.g., Miller et al. (2010) in the refs), then this should be clarified for the readers. If not, then this should be made clear too.

2. Reply:
Two measurement campaigns were conducted, one in late Arctic summer (from the 24th August to the 7th of September, 2009) and one in the late Arctic winter (from the 15th of February to the 7th of March, 2010). Results from the winter campaign are presented in this paper. Although the summer campaign took place before the winter campaign, data were first analyzed for the winter campaign. A manuscript comparing SSA properties from Arctic winter and Arctic summer water together with a more extensive discussion on possible implications for future Arctic climate is in preparation as a separate paper.

2. Revision:
In the manuscript it was written:
“Laboratory experiments using Arctic Ocean sea water were carried out at Ny-Ålesund (78° 55’ N, 11°56’ E), Western Svalbard (Fig. 1a) in a marine laboratory during late Arctic winter

conditions from February to March, 2010. Sea water samples each of 180 l were collected at three different locations in the vicinity of Ny-Ålesund to cover possible differences between outer-fjord and inner-fjord conditions, including the potential influence of the Kongsbreen glacier (Fig. 1b)."(lines 6-11, p 16090)
Some sentences were added:
"Laboratory experiments using Arctic Ocean sea water were carried out at Ny-Ålesund (78° 55′ N, 11°56′ E), Western Svalbard (Fig. 1a) in a marine laboratory during late Arctic winter conditions **(from the 15th of February to the 7th of March 2010) and during late Arctic summer conditions (from the 24th August to the 7th of September 2009)**. **This paper presents results of the winter measurements, whereas Zábori et al. (2012,*in prep*) will compare summer and winter conditions.** Sea water samples each of 180 l were collected at three different locations in the vicinity of Ny-Ålesund to cover possible differences between outer-fjord and inner-fjord conditions, including the potential influence of the Kongsbreen glacier (Fig. 1b)."

3. Comment:
The authors have chosen to work with median as a statistical characteristic instead of average value. Some justification for this choice is necessary. This choice is similar to that used by Hultin et al. (2010), a paper involving most of the co-authors in this manuscript. Figure 3 in Hultin et al. compare average and median size distributions, but, again, the differences and the choice to use median are not discussed. Here the authors have the possibility to make their case. Perhaps additional panel in Figure 3 can show median and average size distributions, and the differences can be quantified (e.g., as percent difference) and used to rationalize the choice to work with the median.

3. Reply:
In the paper by Hultin et al. (2010) we presented both median and mean bubble size distributions, and the difference between them were very small. This is a simple way of showing to the reader that the bubble distribution was not in any significant amount skewed. The same was true for the new data set, and therefore we plotted only the arithmetic mean. We have however now updated the figure and included both mean and median for one exemplarily water type. In addition the percent differences between the arithmetic means and median values were calculated for each bubble size range (and each water type). Overall, the differences between median and mean were small. Because of clarity reasons, only the median of the bubble spectra measured in water sampled outside the fjord is shown. This water type had the largest discrepancy between arithmetic mean and median. The mean of other water types would simply be too difficult to distinguish from the other curves. For the mean curve from water sampled outside the fjord, the difference was with 30 % largest at a bubble radius of 51 µm. For 15 bubble size ranges (out of 20) the discrepancy between the median and arithmetic mean was smaller than 5 % (especially for all bubble diameters larger than 123 µm). For the particle number concentration and particle number size distribution plots, the median was used to describe the population. This was done, as the median is more robust concerning outliers.

3. Revision:
To Fig. 3 (p16121) a median was added, as described above and the figure caption was changed accordingly:

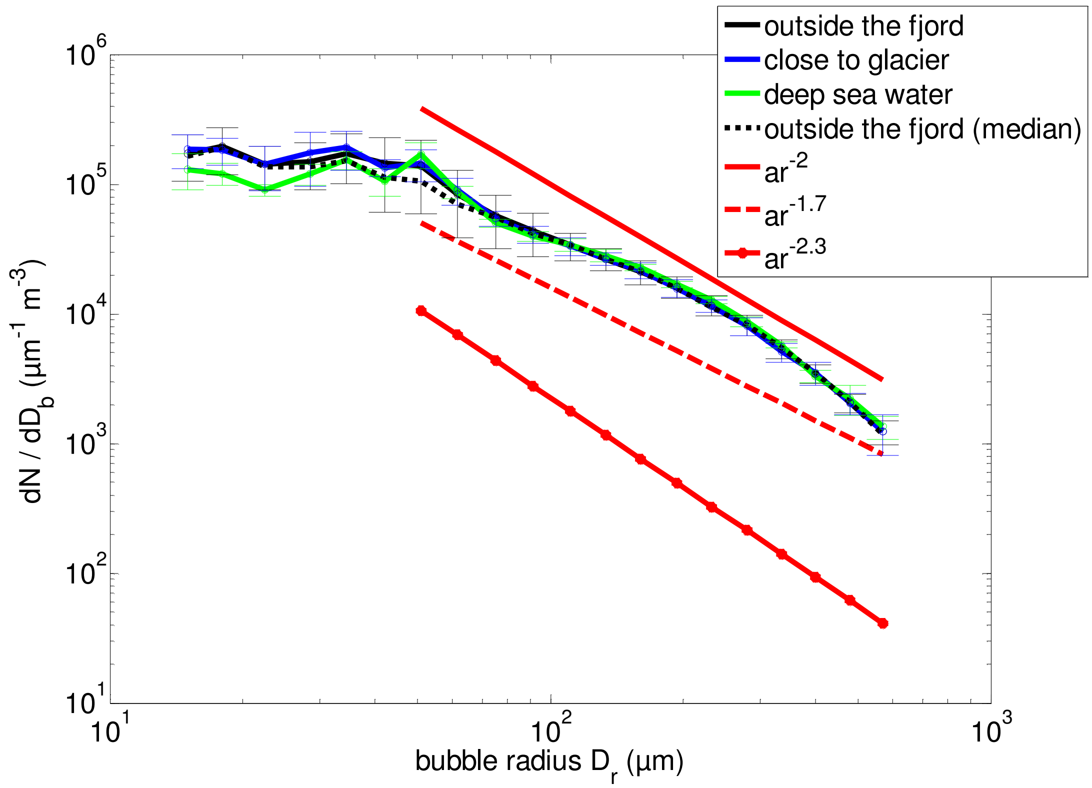

Fig. 3. Solid lines represent arithmetic means with standard deviations for bubble population distributions versus bubble diameter for different water sampling locations. **The black dashed line shows as an example a median for comparison with the arithmetic mean.** Averages are based on 28, 9, and 5 bubble spectra measurements from bubbles produced in water with its origin outside the fjord (black line), close to the glacier (blue line) and deep water (green line), respectively. Red lines represent power law functions dN/dr = ar− b with the bubble radius r for b = 2, b = 1.7 and b = 2.3.

For explaining the presence of the median in Fig.3, some sentences were added after line 2 on page 16097:

"**In addition to the arithmetic mean values of the bubble spectra shown in Fig.3, a median is shown as an example. For the bubble size range which follows the typical power law function of bubbles in the real ocean (Db>0.1 mm), the arithmetic mean and the median of the bubble spectra are essentially not different. Even at sizes Db<0.1 mm, the difference is limited to at the most 30 %. For the other water types, the difference is smaller. "**

4. Comment:

Types of experiments are described in section 3.2 (p. 16097) and elsewhere. Often "single" experiment is mentioned. Only in the end (lines 23-24, 16107) the definition of a "single" experiment is clearly given. Suggest moving the definition "with water sampled." much earlier, say in section 2.5. Clarify "single" versus what other type of experiments–"double"? "multiple"? In other words, consider introducing names and definitions for each experiment and what is expected to get from each experiment as early as possible, and then use these names consistently throughout the text.

4. Reply:
We agree that it makes no sense to use the term “single” experiment. We only conducted one type of experiment: measurements were made during steadily changing water temperature (warming or cooling) during a coherent time period. Differences between the experiments are (except that they are conducted with waters sampled at different locations) that some of them are conducted with water directly after sampling and others are conducted with water which was sampled at the same time, but stored over night. We propose to delete the term “single” and to stress in the text when experiments were conducted with water sampled on the same day, or later after overnight storage.

4. Revision:
The word “single” and “individual” in combination with the word “experiment” was deleted in the manuscript:

a) The sentence:
“The typical duration of a single experiment was approximately 6 h.” (line 15-16, p 16095) was changed to: “The typical duration of **one** experiment was approximately 6 h.”

b) In the sentence:
” The particle number concentrations as a function of Tw between – 2 ℃ and 9 ℃ are shown for each single experiment as a median for each temperature bin in Fig. 4” (line 4-5, p 16097), the word “**single**” was deleted.

c) The caption of Fig. 4 was changed from:
“Medians of particle number concentration for distinct water temperature bins and single experiments.”(p 16122) to
“**Medians of particle number concentration for distinct water temperature bins and each experiment.**”

d) In the sentence:
“For the experiments based on water sampled close to the glacier and deep sea water, the two individual experiments were conducted with water sampled at one single time, but half of the water was stored before starting the second experiment.” (lines 11-14, p 16097) the word “**individual**” was deleted.

e) The sentence:
“The slight difference the magnitude of the peak between the different waters may result from the large variability in particle number concentration between the single experiments (Fig. 4).” (lines 17-20, p 16104) was changed to
“The slight difference **in** the magnitude of the peak between the different waters may result from the large variability in particle number concentration between the experiments (Fig. 4).”

f) The sentence:
“Repeating single warming experiments with water sampled at the same time but used on two different days showed particle number concentration differences of up to 97 % (for the same size range and water temperature).” (lines 23-25, p 16107) was changed to:

“**Conducting** warming experiments with water sampled at the same time but used on two different days showed particle number concentration differences of up to 97 % (for the same size range and water temperature).”

g) The sentence:
“Our results suggest that the organic fraction of the SSA, under the conditions observed, is not controlling the number concentration itself (considering one single experiment).” (lines 17-19, p 16108) was changed to:
“Our results suggest that the organic fraction of the SSA, under the conditions observed, is not controlling the number concentration itself (**within one** experiment).”

5. Comment:
The solubility of oxygen depends on the water temperature. Can the experiments reported here separate the effects of water temperature and oxygen saturation?

5. Reply:
This is a good point. In our experiments, the effect of water temperature and oxygen saturation can indeed be separated for the change of particle number concentration with a change in water temperature. We did so by examining the influence of oxygen saturation on the particle number concentration for the near constant water temperature ranges of 5-6 °C and 1-2 °C (section 3.4 on p 16102). Within these water temperature ranges the water temperature changed continuously and a relationship between water temperature and particle number concentration was observed. The fact that we did not observe any relationship between the oxygen saturation and the particle number concentration for these water temperature bins, showed that it must have been from an oxygen saturation independent water temperature effect.

5. Revision:
Based on the reply above we have added a clarifying text to the section in which the effect of sea water temperature on the particle number concentration is discussed.
Two sentences are added after the discussion that the water temperature dependent trend of particle number concentration was already observed in other studies (lines 2-22, p16107) and before the discrepancy in particle number concentration for experiments on other days is discussed (lines 23 ff, p 16107):
“**We conclude that the observed trend of particle number concentration with water temperature is not due to changes of oxygen saturation. This conclusion is based on the fact that a change in oxygen saturation between 72 % and 83 % for the water temperature range 5-6 °C and 1-2 °C did not cause any change in particle number concentration, whereas a change in water temperature within these small intervals did.**”

6. Comment:
Regarding “hypothesis” in Line 12 (16108)–it would be beneficial for the discussion to formulate this hypothesis early in the text, say in the end of the Introduction. How the current average seawater properties are expected to change as Arctic conditions evolve toward warmer state, e.g., in future warmer conditions on average the seawater temperature would increase, the salinity would decrease, and the organic content would increase. How these new average properties would affect the sea spray production? Then in

the discussion show if your results confirm or repudiate your hypothesis for each of the considered variables.

6. Reply: This comment was very helpful in improving the structure of the manuscript, and we have tried to include the suggestions by the reviewer.

6. Revision:
a) The sentence:
"In this work, the influence of sea water temperature, salinity and oxygen saturation is investigated with respect to primary marine sea spray aerosol emissions." (lines 27-28, p 16089) is replaced by the following sentences:
"**In this work we test the hypothesis that primary marine sea spray aerosol emissions are affected by an on average higher water temperature, lower salinity and a change in an unknown direction of the oxygen saturation (as a result of a change in biological activity).**"
b) The results of testing the hypothesis for the different parameters is proposed to be summarized before the results are discussed in detail. Therefore, the following sentence was added between the chapter caption "Discussion" (line 22, p 16104) and section 4.1 (line 23, p 16104):
"**The hypothesis, that primary marine sea spray aerosol emissions are affected by changed physical properties of the Arctic Ocean is partly confirmed and partly repudiated. The hypothesis was repudiated for a change in oxygen saturation between 72 % and 83 % and could not be confirmed for a change of salinity between 36 ‰ and 26 ‰ (for a wide range of different water temperatures). The hypothesis that an increase in average water temperature impacts on SSA emissions was on the other hand confirmed. The results which led to these conclusions will be discussed separately for the different tested parameters in the following sections, beginning with a discussion of the results regarding how water temperature influenced the air bubble spectra. An influence of water temperature on air bubble spectra was not included in the hypothesis, but was expected to give an explanation for possible observed relationships between the tested physical properties and the SSA emissions.**"

c) To avoid any confusion about the hypothesis of this manuscript, the words "the hypothesis" is deleted in the following sentences:
"However, our observations, and especially the sets of mirroring warming/cooling experiments, support **the hypothesis** that for winter Arctic Ocean seawater, most of the variation in particle number concentration originated from sea water temperature changes and not from a depletion of organic substances from the sea water." (lines 11-14, p 16108)

7. Comment:
In section 5 the authors establish the seawater temperature as the most influential variable when wind speed as a forcing factor is absent. But when inferring the implications of this result for SSA production in future Arctic conditions, one needs to consider ice coverage and water temperature drivers together with the wind speed. To claim the water temperature as the most influential factor, one needs to assume that the change of Arctic conditions to warmer state will not change the average wind speed. If this assumption doesn't hold, the discussion could be extended to consider/conjecture whether the effects of ice shirking (more open water) and warmer waters can compensate for possible decrease of average wind

speed over warmer Arctic. Or masking of the water temperature effect if the average wind increases. What would be the net result of the interplay of these three forcing factors– increased or decreased SSA production? The experiments reported here can't give full answers to such questions, but the questions can be posed.

7. Reply: Following the suggestion by Refree1 and by Referee2 (Comment 5), Section 5 is augmented by a listing of several potential feedbacks which may occur in a future Arctic climate.

7. Revision:
Section 5 (lines 2-16, p 16109) was modified from:
"The observed trend of decreasing SSA production with increasing water temperature may have large implications for the climate in the Arctic region. The diminishing sea ice will result in a decreased surface albedo and contribute to a positive feedback of the Arctic warming. At the same time, larger areas of ice-free ocean will provide large areas of potential SSA emissions, which in turn can act as a negative feedback by increasing aerosol scattering and by modifying cloud microphysical properties providing additional cloud condensation nuclei (cf. Struthers et al., 2011). On the other hand, with increasing sea water temperature and as shown in this study, the sea spray source strength might decrease and thus weaken the negative feedback of SSA on Arctic climate. The magnitude and interplay between the decrease of sea ice coverage and the increasing sea water temperature should be addressed in large-scale model studies, where changes in meteorology, ocean characteristics and marine aerosol emissions all are represented in a consistent manner. A new sea spray aerosol emission parameterization, representing the effects of low sea water temperatures on the SSA emission strength, would be useful to develop for these types of studies."
to:

"The observed trend of decreasing SSA production with increasing water temperature may have large implications for the climate in the Arctic region. The diminishing sea ice will result in a decreased surface albedo and contribute to a positive feedback of the Arctic warming. At the same time, larger areas of ice-free ocean will provide large areas of potential SSA emissions, which in turn can act as a negative feedback by increasing aerosol scattering and by modifying cloud microphysical properties providing additional cloud condensation nuclei (cf. Struthers et al., 2011). On the other hand, with increasing sea water temperature and as shown in this study, the sea spray source strength might decrease and thus weaken the negative feedback of SSA on Arctic climate. **Another important factor influencing the sea spray aerosol emissions is the wind speed. In order to answer questions about how changes in SSA emissions influence the future Arctic climate, it is important to consider all of the above-mentioned factors. To summarize, there are a number of potential feedback processes between a future changing climate, changes in surface albedo and changes in sea spray production, for example:**

- **Increasing (decreasing) water temperature will decrease (increase) sea spray emissions due to changes in the physical properties of water (present study; Bowyer et al., 1990; Hultin et al., 2011).**
- **Increasing (decreasing) wind velocities will result in increased (decreased) sea spray emissions (Lovett, 1978; Nilsson et al., 2001; Geever et al., 2005)**

- **Increasing (decreasing) water temperature will increase (decrease) whitecap fraction and increase (decrease) sea spray emissions (Monahan & O'Muircheartaigh, 1986)**
- **Increasing (decreasing) wind speed will increase (decrease) whitecap fraction and thereby increase (decrease) albedo (Monahan & O'Muircheartaigh, 1986)**
- **Increasing (decreasing) temperature will decrease (increase) sea ice cover and increase (decrease) sea salt emissions (e.g. Nilsson et al., 2001; Struthers et al., 2011).**
- **Increasing (decreasing) temperature will decrease (increase) sea ice cover and decrease (increase) surface albedo.**

Struthers et al. (2011), however, indicated that the impact of future changes in wind speed on the sea salt aerosol production over the Arctic Ocean was small compared to those associated with changes in sea ice coverage and sea surface temperature. All in all, the magnitude and interplay between the decrease of sea ice coverage, the increasing sea water temperature, changes in wind speed and the possible accompanied change in whitecap coverage should be addressed in large-scale model studies, where changes in meteorology, ocean characteristics and marine aerosol emissions all are represented in a consistent manner. An updated sea spray aerosol emission parameterization, which better represents the effects of low sea water temperatures on the SSA emission strength, would be useful to develop for these types of studies."

Technical corrections:

All technical corrections by the reviewer have been taken and the paper has been changed accordingly.

References

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