Response to anonymous Referee#2: Zábori et al., 2012.

The authors thank anonymous referee#2 for insightful comments on the manuscript. The reviewer provided several suggestions for improving the quality of the manuscript. We have considered all comments and suggestions by the reviewer, and our detailed responses are listed below.

1. Comment:
The authors did not measure the partial pressure of dissolved N2 along with dissolved O2. Both are important for bubble formation in weir bubble generation.

1. Reply:
We agree with the reviewer that it would be beneficial to measure N_2 along with the dissolved oxygen, or the total gas pressure. Your comment made clear that we need to explain our choice to measure oxygen saturation. We do state in the manuscript on lines 24-25, p 16089 that "Changing environmental conditions will influence the fauna and flora in the Arctic Ocean […]". The changes in the environmental condition associated with melting sea ice and increasing sea water temperature 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). The three examined physical properties of the Arctic Ocean water (water temperature, salinity, oxygen saturation) were chosen as we expect them to change in future and we had instrumentation available to measure these parameters. We are aware that other parameters, for example N_2 in the ocean will likely change as well, and influence bubble formation. Unfortunately we did not have the available instrumentation to measure N_2 and we focused on the oxygen saturation which is known to vary depending on biological activity and which was shown to impact on the sea spray production (Hultin et al., 2011).

1. Revision:
To clarify why we measured the oxygen saturation, we changed and augmented:
"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 24-25, p 16089)
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:
There should have been some attempt made to constrain the effect of naturally occurring surfactants on bubble populations. If surface tension is an important parameter as the authors claim, surfactants have a much larger potential effect than temperature and the effect should be accounted for, rather than assumed unimportant.

2. Reply:
We do not see the influence of surfactants on bubble formation and particle production as unimportant, but as we had not the necessary equipment to measure the surfactants present, or the surface tension, our experiments were designed to address these open ends indirectly. First of all we believe that we can safely assume that the surfactant concentration did not vary significantly during the course of the campaign. If that would be the case, knowing that surfactants are one of the key players in bubble bursting and consecutive the SSA production process, then we would not get repeatedly the same results and trends. The other option we considered is that surfactants might get depleted during the bubbling process in the laboratory. This option was checked by using the same water in warming and cooling experiments. If significant depletion of the surfactants would occur, we would not have seen the same trend with temperature during both warming and cooling experiments (lines 11-17, p16108).
Furthermore, we do suggest examining the influence of organic matter on physical properties of the water in further studies (lines 19-21, p16108). In another paper by Zábori et al. (2012) the influence of succinic acid, a surfactant which is synthesized and metabolized in different biochemical processes in sea water (Steinberg and Bada, 1984; Kester and Foster, 1963), on the water temperature dependent trend of particle number concentration produced from a NaCl solution was examined (Zábori et al., 2012). Succinic acid was used as a proxy for surface active compounds as it was used previously in aerosol science with respect to aerosol activation in clouds and related changes in surface tension. We agree with the referee that the examination of the bubble spectra for different surfactants and surfactant concentrations is an important contribution to understand the processes driving SSA emissions, but this would exceed the scope of the study.

2. Revision:
We added some sentences which stress that organics were shown to be important for bubble development, although they were not considered to be responsible for the observed temperature dependent trend of particle number concentrations. The sentences:

"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). This hypothesis needs further testing and has to be experimentally explored as other studies have shown an effect of organic matter on physical properties of water which may alter air bubble generation. For example, Nägeli and Schanz (1991) reported that surface tension was reduced by phytoplankton exudates and Lion and Leckie (1981) theoretically described the decrease of surface tension caused by surface-active organics. Garrett (1967) observed a

stabilization of air bubbles on the air-sea interface due to surface-active substances scavenged by the air bubble while rising to the water surface."
Are changed to:
"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). This hypothesis needs further testing and has to be experimentally explored as other studies have shown a **clear** effect of organic matter on physical properties of water which may alter air bubble generation. For example, Nägeli and Schanz (1991) reported that surface tension was reduced by phytoplankton exudates and Lion and Leckie (1981) theoretically described the decrease of surface tension caused by surface-active organics. **An impact of organics on bubble properties was determined by** Garrett (1967). A stabilization of air bubbles on the air-sea interface due to surface-active substances scavenged by the air bubble while rising to the water surface was observed."

3. Comment:
The authors might want to review the modeling work of Jaeglé et al. (2011), who modeled global distributions of sea salt aerosols, including high latitudes.

3. Reply:
We thank the reviewer for pointing out this reference. The work of Jaeglé et al. (2011) also fits very well with the first part of Comment 5a by reviewer 2, which states that "*No discussion of the relationship between water temperature and whitecap coverage*" was made. Thus, the two comments (3 and 5a) will in the following be considered together.
In the real world it is the wind speed causing breaking waves in the ocean and subsequently generating whitecap coverage, thus wind speed is the main parameter in most model parameterizations describing the size dependent sea spray flux to the atmosphere. However, other factors influence whitecap fraction as well, as suggested by the wide variation of whitecap fraction for a given wind speed. Some of them were recently reviewed by de Leeuw et al. (2011). However, in laboratory experiments the whitecap fraction is determined by the way the whitecap is generated. In our laboratory experiments we have no wind speed causing a breaking wave, but we have air entrainment in the water and as a consequence a small whitecap area in the experimental bottle. It was shown in e.g. Monahan & O'Muircheartaigh (1986) that water temperature influences the whitecap coverage for a constant wind speed. Higher water temperatures result in general in larger whitecap coverage. This was mainly attributed to the production of smaller air bubbles in the water with lower terminal rise velocities compared to larger bubbles with an increase in water temperature. In another study it was shown that the time constant characterizing the decay of whitecaps changes inversely with the terminal rise speed of the smaller bubbles (Monahan et al. (1985) in Monahan & O'Muircheartaigh (1986)). Jaeglé et al. (2011) compared globally modeled and observed mass concentrations of coarse mode sea salt aerosol and concluded that the modeled bias was improved when introducing an increased sea salt production with increasing temperatures. As one explanation for the water temperature dependent trend on the mass of coarse mode aerosols, the temperature dependence of the whitecap fraction was named. In our experiments, we can assume that some water temperature influence on the whitecap fraction was present, at least to some extent. What should be discussed is that if the whitecap area in our laboratory experiment is

dependent on water temperature (as suggested in the above-mentioned references) then this should also imply an increase of particles with an increase in water temperature, but we observe the opposite. We suggest to discuss this in section 4.4 (line 1, p 16107) and also to refer to it in Section 5 (line 1, p 16109), please see "5. Revision".

3. Revision:

Based on Reply 3, a discussion about the dependency of whitecap fraction on sea surface temperature and its relevance to our experiments will be added to Section 4.4 after the line 22, p 16107:

"**Although several studies support our results of a decrease in total particle number concentration with an increase in water temperature (see studies mentioned above), there are some studies indicating the opposite relationship. Monahan & O'Muircheartaigh (1986) demonstrated that for a constant wind speed, an increase in water temperature enlarges the whitecap fraction on the ocean surface. This is important, as the sea spray aerosol production is considered proportional to the whitecap fraction. Jaeglé et al. (2011) compared globally modeled and observed mass concentrations of coarse mode sea salt aerosol (in their study taken to be particles with a radius between 0.3 and 3 μm) and concluded that the modeled bias was improved when introducing an increased sea salt production with increasing water temperature. The increase of whitecap fraction with an increase in surface water temperature can be explained by an increased production of smaller air bubbles with slower terminal rise velocities compared to larger bubbles with an increase in water temperature. Monahan et al. (1985, in Monahan & O'Muircheartaigh (1986)) showed that the time constant characterizing the decay of whitecaps changed inversely with the terminal rise speed of the smaller bubbles. Anguelova & Webster (2006) stated that a decrease in viscosity caused by higher water temperatures facilitated wave breaking and as a consequence prolongs the lifetime of a whitecap. Another suggested explanation for observed large-scale increases in whitecap fraction with an increase in water temperature is the difference in the duration of a certain wind speed over different areas. Trade winds, for example, occur over relatively warm waters and persist relatively long so that whitecaps can fully develop, whereas over colder waters the duration of high wind speeds is relatively short (Monahan & O'Muircheartaigh, 1986).**

During our laboratory experiments, we did indeed produce small areas of whitecaps as a consequence of air bubbles reaching the water surface. However, we did not observe any increase in particle number concentration with an increase in water temperature, as one would expect if the whitecap fraction depends on the SST as suggested by Monahan & O'Muircheartaigh (1986) and Jaeglé et al. (2011). One explanation could be that the water surface in the experimental bottle was too limited to allow for an undisturbed whitecap fraction evolution (that wall effects limited the bubble plume and hence the white cap size). On the other hand, no change in the bubble spectrum with temperature was observed either, which is notable as this should be a major cause of the whitecap fraction change with water temperature. Another possible reason for the contradictory results obtained in our study and the ones presented by Jaeglé et al. (2011) could be that the latter focused on coarse mode concentrations of sea salt whereas the temperature dependence observed in our study was most clear for aerosols with a diameter smaller than 1 μm. A positive temperature trend could also be explained by the results of Mårtensson et al. (2003), which in difference to the current study saw increasing aerosol

number produced at diameters larger than about 350 nm with increasing temperature (and decreasing numbers for smaller particles in agreement with the current study)."

4. Comment:
Relative humidity is very much lower than in the tank than in the atmosphere, 10% in the tank versus maybe 50%-60% in the arctic. Since the rate at which the bubble film thins is related to when it breaks and therefore how much aerosol is generated (and the size range of that aerosol), there should be some discussion of how RH is related to the measured size distribution.

4. Reply:
The relative humidity was monitored in the sampling line close to the instruments and not in the bottle where the aerosols were produced. The sentence: "Due to clean air supplied to the tank, the relative humidity during the experiments was always lower than 10 %" (line 19-20, p16093) is misleading. The RH during the experiments in the sampling line was always lower than 10 % and it was not the clean dilution air, but rather the relative high temperature in the sampling line which reduced the RH, compared to the RH above the cold water surface in the bottle where the aerosols were produced.

4. Revision:
Based on Reply 4, the sentences:
"The relative humidity of the sampled air was monitored in the sampling line prior to entering individual instruments with a Hygroclip SC04 hygrometer (Rotronic). Due to the clean air supplied to the tank, the relative humidity during the experiments was always lower than 10 %. Hence, we can safely assume that the observations are representative for dry diameter aerosol particles." (lines 17-21, p) was changed to:
"The relative humidity of the sampled air was monitored in the sampling line prior to entering individual instruments with a Hygroclip SC04 hygrometer (Rotronic). **The relative humidity near the sensing instruments was always lower than 10 %, which is mainly a result of the relative high temperature in the instrument payload. Whereas the aerosols were characterized as dry aerosol particles, the RH conditions in the bottle where the bubbles were produced were significantly higher.**

5. Comment:
a) No discussion of the relationship between water temperature and whitecap coverage,
b) and how changes in whitecap coverage might be of more relevance in a warming arctic than small changes in bubble or aerosol populations.

5. Reply:
a) Please see reply to Comment 3 above.
b) Based on Comment 5 by Referee 2 and based on Comment 7 by Referee 1, Section 5 is augmented by a listing of several potential feedbacks which may occur in a future Arctic climate.

5. Revision:
a) Please see "3. Revision"

b) Section 5 (lines 2-16, p 16109) was changed 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; Mårtensson et al., 2003; 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."

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