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 constructive 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:
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.

1. Reply:
We do not agree with the reviewer's statement that the MQ water has no relevance. The purpose of using MQ water and present the data, was to establish a baseline for the experiment and we consider it as important to show what the baseline is with respect to the rest of the results. The water of all subsequent experiments is based on the same MQ water with added salt or organics. Even if particles are produced by MQ water alone, it is important to show that the contribution is small in comparison to water where NaCl was added. As we required almost 200 L of MQ water for the experiment, it would have been unnecessarily expensive to use ultra pure water. To adhere to some of the criticism we removed Fig. 4 and Table 1 from the manuscript.

1. Revision:
Figure 4 is removed from the manuscript. To make the manuscript consistent, the sentence: "To test the functionality of the experimental setup and to create a baseline case two median size distributions of MQ water (MQ1 and MQ2) were compared to a median size distribution of Arctic Ocean water. The number of particle size distributions used to calculate the respective median is presented in Table 1."(lines 8-11, p19046) was removed. As well as the sentences:
"Median aerosol number size distributions from experiments conducted with MQ water for two different water temperature ranges (3.1 ◦C<T w <3.6 ◦C and 4.8 ◦C<T w <7.4 ◦C) were measured about 4 h apart, and named MQ1 and MQ2, respectively (Fig. 4). As a reference, the two size distributions are compared to a number size distribution of Arctic Ocean water with a similar water temperature (T w = 3.8 ◦C), seen in Fig. 4. Both MQ size distributions show a relatively low number concentration, which is decreasing from Dp 0.01 µm to about Dp 3 µm, while the aerosol size distribution observed for real Arctic Ocean sea water shows an increasing concentration for particles with diameters between Dp 0.01 µm and Dp 0.16 µm. MQ2 shows higher particle concentrations (in total 61 particles cm^{-3} compared to 23 particles cm^{-3} for MQ1), but matches the MQ1 number concentration for particle sizes larger than Dp 0.16 µm."(lines 4-14, p19049)

2. Comment:

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.

2. Reply:

We thank the reviewer to direct our attention to our choice to use a NaCl solution for particle production, which made clear that we did not sufficiently explain our choice of a) the substance we used and b) the concentrations we examined.

Within our experiments we are not aiming to simulate climate change in the Arctic Ocean, but one point is to test the reproducibility of the temperature dependence on particle number concentrations observed in Zábori et al., 2012 and to test how other parameters impact on it (salinity, succinic acid concentration). We are aware that the compounds used (NaCl/succinic acid) and their concentration ranges (salinity range, succinic acid concentration range, but as well the water temperature range), are covering a much wider range than one can expect for Arctic Ocean conditions. We wanted to answer the question, if and to what degree the water temperature dependent trend observed in the Arctic Ocean (Zábori et al., 2012), is influenced by salinity, to make it very clear a salinity of 18 g/kg was chosen. No clear trend of salinity on the particle number concentration could be determined, when comparing particle number concentrations arising from Arctic Ocean water having salinities between 26 and 36 ‰ (Zábori et al., 2012). In the presented experiments (using only NaCl) we tested the influence off different salinities in the absence of any organic compound. The large salinity range was chosen to generally answer the question how salinity is impacting on particle number concentration. To make an impact clear this was done by exaggerated low salinities.

Moreover, one of the outcomes of the field experiments with real Arctic Ocean sea water was the exponential decrease of the PMA emissions with increasing sea water temperature (Zabori et al., 2012). The observations indicate that physical sea water properties are likely responsible for this trend. To exclude the possible influence of organic compounds and surfactants we have chosen NaCl, the purest proxy we had available. For the purpose of this experiment we believe that it fulfilled its role as good as artificial sea salt. Using real sea salt for our experiments we couldn´t exclude that there is a certain amount of organic matter included.

The salinity in the shallow mixed layer of the Arctic Ocean often reaches below 30 g/l during the seasonally repeated melting of sea ice. The most extensive data set is probably from the SHEBA one year ice camp in the late 90's. They frequently observed salinity in the range

lower than 30 ‰ is about as often as the range above 30 ‰ for summer conditions when a net sea ice melt existed (Mcdonald et al., 2002; McPhee et al., 1998). This is confirmed by other separate studies, for example Raskoff et al. (2010). The record reductions in summer sea ice in recent years may very well have resulted in even lower salinity in some areas. At least recent remote sensing data suggest that sea ice melt and changed atmospheric and ocean circulations in combination are building up large amounts of fresh water in the central Arctic Ocean during the summers (Giles et al., 2012). If we widen the perspective somewhat geographically salinities well below 18‰ can be found in large areas outside the Siberian rivers (EWG, 1998; Stepanova et al., 2007), near the outflow of melting glaciers (Nielsen et al., 2010), and of course in brackish seas like the Baltic Sea. Also with a wider time perspective the sea surface salinity have for periods been lower in the Arctic Ocean in past climates (Bauch and Polyakova, 2003). So with a wider perspective, there is clearly a scientific interest also for the behavior of sea spray at salinities below 30‰.
However, due to Comment 2, the size distributions based on 30 - 31 ‰, 32 - 33 ‰, 33 - 34 ‰ and 34 - 35 ‰ salinity water were compared. For the range 31 – 32 ‰ too few data were available. The comparison did not show any systematic difference in number size distribution between different salinities.

2. Revision:
Obviously the aim of our study was not stressed sufficiently. Therefore, the paragraph: "However, in this study we focus on the effect on NaCl particle generation from bubble bursting caused by changes in water temperature near freezing conditions, motivated by observations of Arctic Ocean water (Zábori et al., 2012) which revealed a strong temperature dependence of particle emissions near freezing temperatures. We also investigate the influence of salinity and surface-active compounds represented by succinic acid on total aerosol numbers and number size distributions. The objective of this work, combined with laboratory experiments conducted with real Arctic sea water, is to quantify the relative importance of water temperature, salinity, and other dissolved species in influencing primary marine aerosol production in high latitude ocean waters. This might support parameter considerations for further aerosol emission parametrizations for this region."(lines 27-30, p19041 and lines 1-8, p19042) was changed to:

"In this study we focus on the effect on NaCl particle generation from bubble bursting caused by changes in water temperature near freezing conditions, motivated by observations of Arctic Ocean water (Zábori et al., 2012) which revealed a strong temperature dependence of particle emissions near freezing temperatures. **The impact of salinity and a surface-active compound (succinic acid) on the water temperature dependent trend of particle number concentration is studied by varying NaCl and succinic acid concentrations on a wider range than expected in real conditions. This is to test how robust and influential links between salinity, surfactant concentrations and PMA emissions are. Through these experiments we test the hypothesis indicated in Zabori et al., 2012 that changes in PMA emission from water with temperatures close to the freezing point are likely controlled by sea water physical properties without a significant impact of organic compounds.**"

To make clear that we want to test the reproducibility of the water temperature dependent trend of particle number concentration observed with Arctic water and that we don´t aim to simulate climate change in the Arctic, the sentence:

“Experiments were conducted for low water temperatures in order to emulate Arctic conditions and to investigate the large sensitivity of particle number concentration expected at low temperatures (Zábori et al., 2012).” (lines4-7, p19045) was changed to:
“Experiments were conducted for low water temperatures in order to emulate **experiments conducted with Arctic Ocean water** and to investigate the large sensitivity of particle number concentration expected at low temperatures (Zábori et al., 2012).”

After the sentences:
“Relative particle number concentrations for smaller and larger sizes differ as well for different salinities, but not in an identifiable direction. One exception is an indication of a weak tail of smaller particles from about 15 g kg^{-1}. Generally, it can be said that there is a local maxima for all salinity bins at around 0.225 µm and the number size distributions do not vary considerably in relative shape for salinities higher than 15 g kg^{-1}” (lines 1-5, p19052) the results of the comparison between particle number size distributions resulting from 30 - 31 ‰, 32 - 33 ‰, 33 - 34 ‰ and 34 - 35 ‰ saline water are added:
“**A comparison of particle number size distributions (not shown) resulting from waters having salinities between 30 and 35 g kg^{-1} (salinity bin size was 1 g kg^{-1} and the salinity range between 31 and 32 g kg^{-1} is not considered due to a small number of observations), did not show any systematic difference in number size distribution between the different salinity ranges. Parameters of log-normally fitted size distributions are listed in Table 3.**”

In addition to give some more information about salinity ranges observed in the Arctic Ocean, some sentences are added to the Introduction. After the sentence:
“Concurrently, salinity in the surface waters is expect to decrease locally, given an expected enhancement in the negative mass balance of glaciers (Nuth et al., 2010).”(lines 10-12, p 19041), some sentences are added:
“**Additionally to the fresh water inflow due to the melt of glaciers, it was indicated that salinities in the Arctic Ocean were lowered by changes in the interplay between wind fields, melting sea ice and river runoff (Giles et al. 2012; MacDonald, 2002). Salinities lower than 30 ‰ are frequently observed during summertime in the surface waters of the Arctic Ocean (Raskoff et al., 2010; EWG, 1998; Giles et al. 2012; MacDonald, 2002; Stepanova et al., 2007).**”

3. Comment:
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.

3. Reply:
The authors are aware that using succinic acid and using higher concentrations than about 0.1 µM are unrealistic for the aim to simulate in-situ changes in surfactant concentrations in the ocean. But, our aim was to test the impact (by adding a surfactant) on the water temperature dependent trend of particle number concentration. By exaggerating concentrations helps us to clarify the relative importance of a change in water temperature compared to changes in other parameters. The highest succinic acid concentration was used in the cooling/warming experiment (Fig. 2 and Fig. 3) as this concentration also had the largest effect on the number of particles generated by the experimental setup. It was never the intention to actually simulate in-situ conditions and disagree with the reviewer's statement that our usage of succinic acid has no scientific values. Just as turning processes on or off in numerical models, using exaggerated or simplified systems in the laboratory helps understanding the real and much more complicated system. Selecting any one surfactant could receive similar criticism as that given in the reviewer's comment simply because it is not real ocean conditions. This includes using a soup as by no means makes our "jar" experiment any more realistic, nor simulates all Arctic ocean regions.

3. Revision:
To clarify, why the highest succinic acid concentration was used when conducting the cooling/warming experiments, the sentences:
" In a final experiment the impact of an organic acid on the temperature dependent trend of particle number characteristics was examined. To achieve this, water with a stable salinity of 35 g kg^{-1} was cooled down from 20.4 ◦C to about 0 ◦C. While keeping the water temperature close to the freezing point, succinic acid (ACS reagent,>99.0 %, Sigma-Aldrich) was stepwise added to the water in the tank, starting with a concentration of 94 µmol l^{-1} which is within the same order of magnitude of the estimated dissolved organic carbon concentration in Arctic Ocean waters (Kivimäe et al., 2010). After a succinic acid concentration of about 2450 µmol l^{-1} was reached, the water sample was left to warm up to a water temperature of about 13.3 ◦C." (lines 27-29, p 19045 and lines 1-6, p19046), were augmented by:
"**It was chosen to conduct the cooling/warming experiment with the highest succinic acid concentration as this had proven to also have the largest effect on particle production.**

To specify the particle number concentration change with a change in succinic acid concentration, the sentences:
"The succinic acid concentration of 94 µmol l^{-1} is the same order of magnitude as the estimated dissolved organic carbon concentration in the Arctic Ocean (Kivimäe et al., 2010) and four orders of magnitude higher than the observed concentrations of succinic acid in the ocean (Tedetti et al., 2006). A decrease of particle number concentration with increasing succinic acid concentration is observed for particles 0.025 µm<D p <0.300 µm. Compared to the number size distribution based on pure NaCl water, the size distributions resulting from water with a 94 µmol l^{-1} and 2446 µmol l^{-1} succinic acid concentration show a 9 % and 43 %

reduced particle number concentration for D p <0.312 µm." (lines 19-27, p 19053) are replaced by:
"The succinic acid concentration of 94 µmol l^{-1} is the same order of magnitude as the estimated dissolved organic carbon concentration in the Arctic Ocean (Kivimäe et al., 2010) and four orders of magnitude higher than the observed concentrations of succinic acid in the ocean (Tedetti et al., 2006). **A change in succinic acid concentration from 0 to 94 µmol l^{-1}, resulted in an about 10 % decrease for all examined sizes. The particle number concentration systematically decreased with increases in succinic acid concentration. Hence, the highest succinic acid concentration tested (2446 µmol l^{-1}) gave the largest result with an average decrease of 43 % for particles smaller than 0.316 µm and in an average decrease of 22 % for particles between 0.316 µm and 4.5 µm. Averaged over all sizes the decrease was about 37 %.**"

The abstract was according to Comment 3 changed from:
"An experiment where succinic acid was added to a NaCl water solution showed, that the number concentration of particles with D p < 0.312 µm decreased by 43 % when the succinic acid concentration in NaCl water at T w = 0 ◦C was increased from 0 µmol l^{-1} to 2446 µmol l^{-1}."(lines 14-17, p 19040) to:
"**An experiment where succinic acid was added to a NaCl water solution showed, that, the number concentration of particles with 0.010 µm <Dp < 4.5 µm decreased on average by 10 % when the succinic acid concentration in NaCl water at a water temperature of 0 ◦C was increased from 0 µmol l^{-1} to 94 µmol l^{-1}.**"

One bullet in the summary was changed from:
"An increase of succinic acid concentration from 0 µmol l^{-1} to 2446 µmol l^{-1} in four steps at T_w = 0◦C tends to decrease the particle number concentration for particles with 0.01 µm<Dp <0.312 µm by about 43 %." (lines 17-19, p19066) to
"**An increase of succinic acid concentration from 0 µmol l^{-1} to 94 µmol l^{-1} at a water temperature of 0◦C tends to decrease the particle number concentration for particles with 0.01 µm<Dp <4.5 µm by about 10 %.**"

In the beginning of the "Discussion and Conclusion" chapter the results and the relevance of the water temperature dependent trend are pointed out. Some sentences are added below line 17, p 19054:
"**Warming/cooling experiments with a NaCl solution for water temperatures between 0 and 21 °C confirmed the water temperature dependent trend of particle number concentration observed for Arctic Ocean water. It was shown, that this trend was clear and reproducible. This was tested by using a NaCl solution with a salinity of 35 g kg^{-1}, an exaggerated low salinity of 18 g kg^{-1} and by using a NaCl solution with an exaggerated high succinic acid concentration of 2446 µmol l^{-1}. Further experiments were conducted with a stable water temperature: It was shown that salinity concentrations between 15 and 35 g kg^{-1} did not impact on the relative shape of the size distribution; within the salinity range 30 – 35 g kg^{-1} no salinity dependent significant difference on the size dependent particle number concentration could be detected; examining six different succinic acid concentrations in the water (between 0 and 2446µmol l^{-1}) showed a decrease in particle number concentration with an increase in the succinic acid concentration.** "

4. Comment:
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.

4. Reply:
We do not agree with the Referee that the total particle number concentration dependence should be presented as a summary. We think that it contains valuable information which is easy to understand with the graphical format chosen: The robustness of the water temperature dependent trend of particle number concentration although an exaggerated low salinity or an exaggerated high succinic acid concentration is present (please see Replies and Revisions to Comment 2 & 3). Tables with log-normal fitting parameters are added to the manuscript, which is often the format preferred by modelers (please see 5.b Reply and Revision).
The Referee refers to Fig. 7 (p 19078) and points out the different concentrations of small particles (10 nm – 20 nm) in the real oceanic environment and within our presented experiments (especially for 12 – 18 g kg^{-1}). We do not completely follow the reviewer´s statement that we considered the particles in the size range above 0.25um as negligible and that we concluded that therefore, DMPS and CPC must agree well. The DMPS range does not cover the whole PMA size distribution. Thus comparing the total CPC with the integral number densities derived from aerosol size distributions will result in higher values of the total CPC, which is an expected outcome. However, we compared the integrated particle number concentrations on which Fig. 7 is based with the total particle number concentration of the CPC. The particle number concentration is 17 % lower for the DMPS measurements compared to the CPC measurements. We do not agree with the Referee that Fig.7 is only useful as supplementary material. As stated above (please see Reply 2) we are not only interested in salinities occurring in the ocean, but we would like to examine the impact on salinity in general and in combination with the water temperature dependent trend in particle number concentration. In addition, in our opinion the evolution of the relative shape dependent on salinity is valuable information, especially as no change in the relative shape is observed for salinities between 15 and 35 g kg^{-1}. However, we followed the Referee´s suggestion to examine the size distributions resulting from waters having salinities between 30 and 35 g kg^{-1}, in more detail (please see Reply 2 and Reply 5b).

5. Comment:
a*) Tables 1-5 can be incorporated into the text as a technical information.*

b) *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.*

5. Reply
a) Tables are deleted and the information is included into the text.
b) Three more tables are added to the manuscript:

1. A table with modal parameters resulting from fitting log-normal functions to the particle number size distributions based on waters having water temperatures of 0 °C, 1 – 4 °C, 8 – 11 °C and 13 – 16 °C (original Fig. 6).

2. A table with modal parameters resulting from fitting log-normal functions to the particle number size distributions based on waters having salinities between 30 - 31 g kg^{-1}, 32 – 33 g kg^{-1}, 33 – 34 g kg^{-1} and 34 – 35 g kg^{-1} (figure not shown in the manuscript).

3. A table with modal parameters resulting from fitting log-normal functions to the particle number size distributions based on waters having succinic acid concentrations of 0 µmol l^{-1}, 94 µmol l^{-1}, 565 µmol l^{-1}, 1505 µmol l^{-1} and 2446 µmol l^{-1} (original Fig. 9).

5a) Revision

Table 1 (p 19082):
is omitted based on Comment 1.

Table 2 (p 19083):
is deleted and the information is incorporated into the text :
"Before the experiment, the frozen MQ water was left to melt until it reached a water temperature of 7.4 ◦C. During the experiment the MQ water was then cooled down to 1.7 ◦C **(cooling rate of 1.1°C h^{-1})** and again left to warm up to 7.4 ◦C **(warming rate of 0.4 °C h^{-1})**."(lines 25-26, p19044 & lines1-2, p 19045)
Information is as well added to lines 13-18, p 19045:
"The experiment using water with a salinity of 18 g kg^{-1} initially had a water temperature of 21.4 ◦C and was cooled down to 0.3 ◦C **(cooling rate of 1.2°C h^{-1})** before the water was left to warm up to 12.6 ◦C **(warming rate of 0.8°C h^{-1})**. For the 35 g kg^{-1} salinity water (representing typical ocean salinity) the corresponding temperatures were 16.8 ◦C at the start of the experiment, 0 ◦C was the lowest temperature and 14.2 ◦C at the end of the experiment. **The corresponding cooling and warming rates were 1.4°C h^{-1} and 1.1°C h^{-1}, respectively.**"

And at lines 28-29, p19045:
"To achieve this water with a stable salinity of 35 g kg^{-1} was cooled down from 20.4 ◦C to about 0 ◦C **(cooling rate of 1.5°C h^{-1})**."

And at lines 5-6, p19046:

"After a succinic acid concentration of about 2450 µmol l^{-1} was reached, the water sample was left to warm up to a water temperature of about 13.3 ◦C **(warming rate 1.1°C h^{-1})**."

To make the text more consistent, the sentence in lines 16-18, p19046:
"On average, cooling water temperature down to the lowest temperature took 12 h 10 min whereas the following warming up time was on average 14 h 20 min." was moved behind lines 5-6, p19046:
"After a succinic acid concentration of about 2450 µmol l^{-1} was reached, the water sample was left to warm up to a water temperature of about 13.3 ◦C **(warming rate 1.1°C h^{-1})**. It was chosen to conduct the cooling/warming experiment with the highest succinic acid concentration as this had proven to also have the largest effect on particle production. On average **for all experiments**, cooling water temperature down to the lowest temperature took 12 h 10 min whereas the following warming up time was on average 14 h 20 min."

The second sentence in lines 18-19, p19046:" The cooling and warming rates for the different experiments are shown in Table 2." was deleted.

Table 3 (p 19084):
Is deleted and its information is incorporated into the text. Instead incorporating the numbers of size distributions, the total length of the measurements for the assigned temperature ranges will be included. Showing the information in this format is more convenient for the reader, as otherwise two numbers for two different measurement ranges has to be named. The text in lines 21-25, p 19046 is modified:
"The first range contained all size distributions resulting from waters having a temperature of 0 ◦C **(total measurement time 1h 25 min)**, the second range included all size distributions between 1 ◦C and 4 ◦C **(total measurement time 6h 20 min)**, the third contained all size distributions resulting from water with Tw between 8 ◦C and 11 ◦C **(total measurement time 3h 30 min)** and the last temperature range included waters with a temperature between 13 ◦C and 16 ◦C **(total measurement time 1h 50 min)**."

The sentence in lines 7-8, p19047: "The number of size distributions used to calculate the median particle number size distribution is shown in Table 3." was deleted.

Table 4 (p 19085):
Is deleted and its information is as the total measurement time incorporated into the text. Please see the motivation above ("Table 3"). The text in lines 17-22, p 19047 is modified:
"The median particle number size distributions resulting from 35 g kg^{-1} NaCl water with different succinic acid concentrations (0 µmol l^{-1}, 94 µmol l^{-1}, 565 µmol l^{-1}, 1505 µmol l^{-1} and 2446 µmol l^{-1}) were compared to test the influence of different succinic acid concentrations in the water on particle number size distributions. **Total measurement times of the size distributions were 50 min for 0 µmol l^{-1} water, 1h 35 min for 94 µmol l^{-1} water, 45 min for 565 µmol l^{-1} water, 1h 10 min for 1505 µmol l^{-1} water and 1h for 2446 µmol l^{-1} water.**"

The sentence lines 20-22, p19047: "Table 4 details the number of size distributions used to build the median for the different succinic acid concentrations." was deleted

Table 5 (p 19086):
Is deleted and its information is as the total measurement time incorporated into the text. Please see the motivation above ("Table 3"). The text in lines 23-26, p 19047 is modified: "To explore the effect of possible additional organics and other inorganic salts than NaCl on particle number size distribution, median number size distributions of unadulterated Arctic water **(total measurement time 25 min)**, NaCl water with a succinic acid concentration of 2446 µmol l^{-1} **(total measurement time 1h)** and of 0 µmol l^{-1} **(total measurement time 1h 10 min)** were calculated."

The sentence lines 27-29, p19047: "The number of distributions used to calculate the medians in different size ranges are summarized in Table 5." was deleted.

5b) Revision

Based on Reply 5b) three more tables with their caption are added to the manuscript:

Table 2: Parameters for fitted lognormal aerosol number size distributions with Nx as the number concentration (cm^{-3}), Dgx as the geometric diameter (µm) and σx as the standard deviation for the different modes and water temperatures (T_w): 0 °C, 1 – 4 °C, 8 – 11 °C and 13 – 16 °C.

T_w	0 °C	1 – 4 °C	8 – 11 °C	13 – 16 °C
N1	**380**	**220**	**60**	**45**
Dg1	**0.037**	**0.035**	**0.03**	**0.028**
σ1	**1.6**	**1.6**	**1.8**	**1.8**
N2	**880**	**540**	**80**	**80**
Dg2	**0.11**	**0.11**	**0.10**	**0.09**
σ2	**1.6**	**1.6**	**1.8**	**1.8**
N3	**770**	**450**	**60**	**50**
Dg3	**0.235**	**0.235**	**0.250**	**0.250**
σ3	**1.25**	**1.25**	**1.25**	**1.25**
N4	**180**	**150**	**38**	**35**
Dg4	**0.52**	**0.52**	**0.58**	**0.58**
σ4	**1.4**	**1.4**	**1.4**	**1.4**
N5	**16**	**18**	**10**	**10**
Dg5	**1.45**	**1.55**	**1.7**	**1.8**
σ5	**1.4**	**1.4**	**1.4**	**1.4**

To refer to the table in the text after the sentences:
"Comparing particle number size distributions of 35 g kg− 1 NaCl water of the four water temperature ranges 0 ◦C, 1 ◦C to 4 ◦C, 8 ◦C to 11 ◦C and 13 ◦C to 16 ◦C show that the number size distribution resulting from the two lower temperature ranges have significant higher particle number concentrations than the size distributions based on the two higher

temperature ranges for particle sizes 0.012 µm<D p <1.5 µm (see Fig. 6). All four distributions overlap for dry diameters outside this range. The two number size distributions resulting from higher water temperatures (T w = 8–11 ◦C and T w = 13–16 ◦C) overlap over the whole size range. A local maximum between D p 0.2 µm and 0.25 µm can be identified for all four size distributions." (lines14-22, p 19051) one sentence is added:

"**Log-normal fittings were performed for all four every size distribution and the resulting number size distribution parameters are listed in Table 2.**"

Table 3. Parameters for fitted lognormal aerosol number size distributions with Nx as the number concentration (cm^{-3}), Dgx as the geometric diameter (µm) and σx as the standard deviation for the different modes and salinities: 30 – 31 g kg^{-1}, 32 – 33 g kg^{-1}, 33 – 34 g kg^{-1} and 34 – 35 g kg^{-1}.

salinity	30 -31 g kg^{-1}	32 -33 g kg^{-1}	33 -34 g kg^{-1}	34 -35 g kg^{-1}
N1	50	55	60	50
Dg1	0.030	0.033	0.033	0.030
σ1	1.55	1.6	1.6	1.6
N2	75	70	70	80
Dg2	0.1	0.1	0.1	0.1
σ2	1.60	1.60	1.60	1.60
N3	45	45	45	45
Dg3	0.24	0.24	0.25	0.25
σ3	1.20	1.20	1.20	1.20
N4	40	40	40	40
Dg4	0.58	0.58	0.58	0.58
σ4	1.4	1.4	1.4	1.4
N5	11	11	11	11
Dg5	1.9	1.9	1.9	1.9
σ5	1.4	1.4	1.4	1.4

To refer to the table after the sentences:

"Relative particle number concentrations for smaller and larger sizes differ as well for different salinities, but not in an identifiable direction. One exception is an indication of a weak tail of smaller particles from about 15 g kg^{-1}. Generally, it can be said that there is a local maxima for all salinity bins at around 0.225 µm and the number size distributions do not vary considerably in relative shape for salinities higher than 15 g kg^{-1}" (lines 1-5, p19052) the results of the comparison between particle number size distributions resulting from 30 - 31 ‰, 32 - 33 ‰, 33 - 34 ‰ and 34 - 35 ‰ saline water are added:

"**A comparison of particle number size distributions (not shown) resulting from waters having salinities between 30 and 35 g kg^{-1} (salinity bin size was 1 g kg^{-1} and the salinity range between 31 and 32 g kg^{-1} is not considered due to data limitations), did not show any systematic difference in number size distribution between the different salinity ranges. Parameters of log-normally fitted size distributions are listed in Table 3.**"

Table 4: Parameters for fitted lognormal aerosol number size distributions with Nx as the number concentration (cm^{-3}), Dgx as the geometric diameter (µm) and σx as the standard deviation for the different modes and succinic acid concentrations in the water: 0 µmol l^{-1}, 94 µmol l^{-1}, 565 µmol l^{-1}, 1505 µmol l^{-1} and 2446 µmol l^{-1}.

succinic acid concentration	0 µmol l^{-1}	94 µmol l^{-1}	565 µmol l^{-1}	1505 µmol l^{-1}	2446 µmol l^{-1}
N1	300	260	240	210	190
Dg1	0.04	0.04	0.04	0.04	0.04
σ1	1.6	1.6	1.6	1.6	1.6
N2	740	680	620	440	360
Dg2	0.105	0.105	0.105	0.105	0.11
σ2	1.6	1.6	1.6	1.6	1.6
N3	720	640	600	540	460
Dg3	0.235	0.240	0.235	0.235	0.235
σ3	1.3	1.3	1.3	1.3	1.25
N4	170	160	150	150	140
Dg4	0.530	0.530	0.530	0.530	0.530
σ4	1.4	1.4	1.4	1.4	1.4
N5	22	16	17	17	17
Dg5	1.55	1.55	1.55	1.55	1.55
σ5	1.4	1.4	1.4	1.4	1.4

To refer to the table the sentences:
"Figure 9 shows particle number size distributions resulting from NaCl waters to which different amounts of succinic acid were added, resulting in succinic acid concentrations of 94 µmol l^{-1} , 565 µmol l^{-1} , 1505 µmol l^{-1} and 2446 µmol l^{-1} in the water. As a reference, a size distribution produced from pure NaCl water is shown in addition. All median water temperatures were in the range between -0.2 and +0.2 ◦C and medians of salinity were in the range between 35.4 and 35.7 g kg^{-1}. The succinic acid concentration of 94 µmol l^{-1} is the same order of magnitude as the estimated dissolved organic carbon concentration in the Arctic Ocean (Kivimäe et al., 2010) and four orders of magnitude higher than the observed concentrations of succinic acid in the ocean (Tedetti et al., 2006). A decrease of particle number concentration with increasing succinic acid concentration is observed for particles 0.025 µm<D p <0.300 µm. Compared to the number size distribution based on pure NaCl water, the size distributions resulting from water with a 94 µmol l^{-1} and 2446 µmol l^{-1} succinic acid concentration show a 9 % and 43 % reduced particle number concentration for D p <0.312 µm." (lines 19-27, p 19053) are replaced and augmented by:

"Figure 9 shows particle number size distributions resulting from NaCl waters to which different amounts of succinic acid were added, resulting in succinic acid concentrations of 94 µmol l^{-1} , 565 µmol l^{-1} , 1505 µmol l^{-1} and 2446 µmol l^{-1} in the water. As a reference, a size distribution produced from pure NaCl water is shown in addition. All median water temperatures were in the range between -0.2 and +0.2 ◦C and medians of salinity were in the range between 35.4 and 35.7 g kg^{-1}. The succinic acid concentration of 94 µmol l^{-1} is the same order of magnitude as the estimated dissolved organic carbon concentration in the Arctic Ocean (Kivimäe et al., 2010) and four orders of magnitude higher than the observed concentrations of succinic acid in the ocean (Tedetti et al., 2006). **A change in succinic acid concentration from 0 to 94 µmol l^{-1}, resulted in an about 10 % decrease for all examined sizes. The particle number concentration systematically decreased with increases in succinic acid concentration. Hence, the highest succinic acid concentration tested (2446 µmol l^{-1}) gave the largest result with an average decrease of 43 % for particles smaller than 0.316 µm and in an average decrease of 22 % for particles between 0.316 µm and 4.5 µm. Averaged over all sizes the decrease was about 37 %. Log-normal fittings to all size distributions shown in Fig. 8 were performed. The resulting number size distribution parameters for all succinic acid concentrations are presented in Table 4.**"

Based on all deleted and all new included tables, Table 6 (p 19087) is renamed to **"Table 1"**. As a consequence, the sentence: "Parameters for the fitted size distributions are listed in Table 6."(line 2-3, p19048) is changed to:
"Parameters for the fitted size distributions are listed in **Table 1**."

6. Comment:
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.

6.Reply:
The presented Arctic Ocean data in Fig.2 and Fig.3 are based on laboratory experiments in the field (with a similar set up presented in the paper) with water collected during two campaigns, one during Arctic summer and one during Arctic winter. Water temperature ranges from -1 to 5°C result exclusively from water sampled during the winter period and water temperature ranges between 10 and 15°C result exclusively from water sampled during the summer period. The temperature ranges in between are covered from both data sets. In total water was sampled from 4 different locations at two different seasons. The Arctic Ocean data in Fig. 2 and Fig. 3 shows medians of particle number concentrations for each sampling location. It is likely that particle number concentrations are scattered within one water temperature range, as each sampling location/time did present different environmental conditions (close to a glacier, summer; close to a glacier, winter; deep water sample, summer; deep water sample, winter; about 10km from glacier away, summer; highly influenced by open waters, summer; higher influenced by open water, winter). Repeating experiments conducted from water sampled at the same location, but at different times showed as well a wide scattering within the same water temperature range. It is likely that the water sampled at different times had different chemical/biological composition. Experiments conducted on two different days with water sampled at the same time and location, showed a wide scattering of particle number concentration for the same water temperature range, too. In between the experiments the water was stored in a dark room at 4°C. Here, a change in biological content in the water cannot be excluded, which might have led to the observed difference.

The absolute concentrations of the Arctic Ocean water cannot be compared to the 35 g/kg data (and any other kind of NaCl experiment), as the dilution rate of the bubble bottle with the clean particle free air differed between the Arctic Ocean and the NaCl experiments. Of course it is possible to adjust the numbers, but this would not clarify the information we are aiming at: We are interested in the rate of change in the particle number concentration with water temperature and want to examine if a pure NaCl solution shows the same trend, despite it is free of organics.

If we adjust the particle numbers of the Arctic Ocean water to the dilution rate of the NaCl experiments, we would get particle number concentrations up to 7000 particles per cm^3 for particles with Dp> 0.01µm and concentrations up to about 2000 particles per cm^3 for particles with Dp> 0.250µm. This would result in a very stretched Fig. 2 and Fig. 3 and would obscure the most important thing we would like to show with the figures: The decreasing trend of particle number concentration with an increase in water temperature for the NaCl solutions.

6. Revision:

Based on Comment 6 we would like to add information about the Arctic Ocean water experiments. Therefore a paragraph is added after line 6, p 19046, which is describing the origin of the Arctic Ocean water data:

“**Even though the focus of this paper is on the experiments conducted with NaCl solutions, data resulting from measurements with Arctic Ocean water are shown in addition. This is because the starting point for the presented study is the observations which were made with Arctic Ocean sea water. Experiments with Arctic Ocean sea water were conducted with a similar set up and instrumentation as described in Section 2.1, 2.2 and 2.4. . Water was sampled during late Arctic summer (from the 24th August to the 7th of September 2009) and late Arctic winter (from the 15th of February to the 7th of March 2010) from in total 4 different locations, in the vicinity of Kongsfjord/Svalbard. Characteristic of the different sampling location was the different influence with glacial melt water. Nevertheless presented data here include only waters with salinities between 34 and 35‰. The sampling during different seasons took place to account for both, biologically more active and less active waters.**”

Further on sentences were added after: ” The particle number concentrations for waters with salinities of about 18 g kg− 1 and 35 g kg− 1 are shown as a function of water temperature (Fig. 2 and Fig. 3), and for comparison particle number concentrations resulting from Arctic Ocean water were added.”(line 7-10, p 19050)

“**The Arctic Ocean particle number concentrations are medians over each measured temperature bin and for every sampling location during the different seasons (late Arctic winter or late Arctic summer)**.”

To refer to the scattering in the Arctic Ocean data after the sentences:

“The data resulting from Arctic Ocean water experiments exhibit the same trend for the particles Dp >0.25 µm albeit with a steeper slope.” (line 26-28, p 19050) it is added:

“**The scattering of the particle number concentration resulting from Arctic Ocean water within the same water temperature range or in neighboring temperature bins is likely a result of the natural variability as the samples were collected at different locations and at**

different times. However, the water temperature dependent trend is consistent for all waters."

7. Comment:
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.

7. Reply:
The choice of the sizes used for building the ratio is based on the instrument characteristics of the DMA and OPC. Authors are aware that using different size ranges would alter the ratio. However, we think that the size intervals used in Fig. 5 serve the purpose of showing that small particles are relatively more important for the water temperature dependent trend. The trend is more easily visible in the ratio figure than in the figure showing size distributions for different water temperature ranges (Fig.6). Therefore, we prefer to keep Fig. 5 in the manuscript.

8. Comment:
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.

8. Reply:
It is true that the presented study differs from several studies as the main sea spray mode is at about Dp 200 nm and not at about 100 nm, as reported. However, we can exclude that a RH above the efflorescence point for NaCl caused the mode at about 200 nm. We wrote "However, since relative humidity was always lower than 45% during the experiments, all particle sizes are referred to as dry diameters" (line 21-22, p19042). This is misleading. During all experiments except one the RH mean was lower than 30 %. The exception is the experiment when salinity was increased from 0 to 18 g kg^{-1}. There the RH mean was 43 % and for this case we cannot exclude that the particle size was influenced by the RH. For Fig. 7

this implies that the number concentrations are most probably shifted somewhat towards smaller sizes for dry conditions.

We are aware that measurements with a fully dried sea salt (RH ~ 10%) would be adequate. Unfortunately, due to a limitation of resources and time, it is not possible for us to repeat the experimental set-up, as recommended. Measurements not presented in the study support our statement that the peak at about 200 nm is not due to the presence of water vapor: Parallel to the size distributions presented in the paper, measurements of the same aerosol population (splitted sampling line) were made passing a thermodenuder having a temperature of 300°C. The resulting size distributions showed as well a major peak at about 200 nm (see Fig.1).

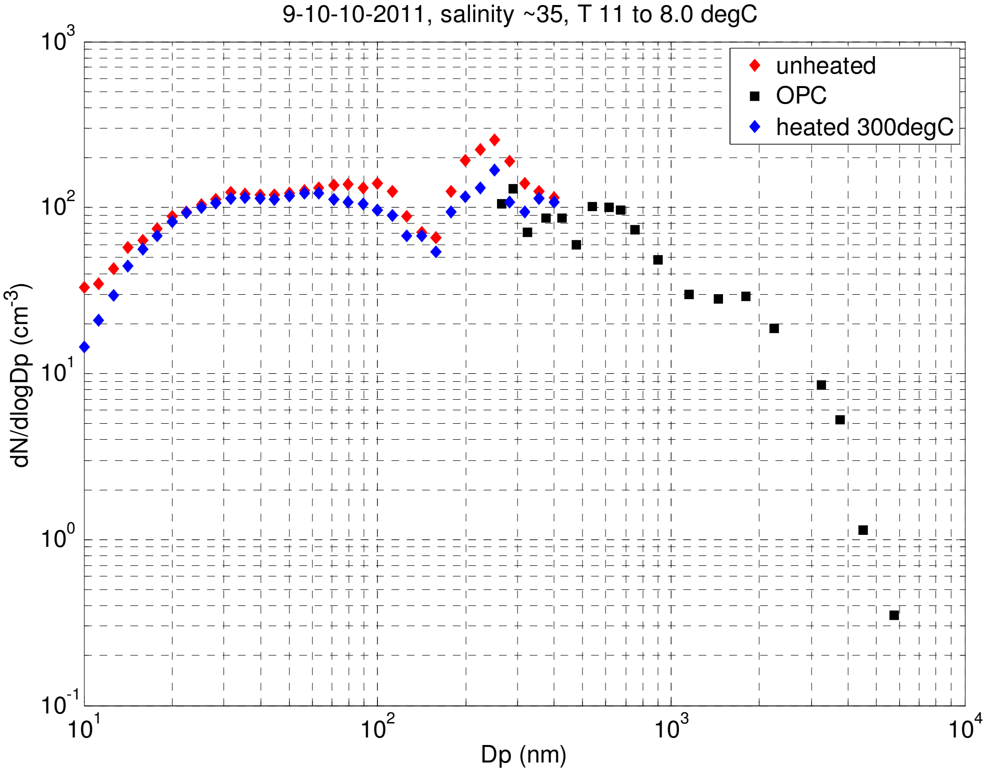


Figure 1: Comparison of an unheated and 300 degC heated size distribution.

Even though many studies report a main sea spray modes at around 100 nm, we would like to refer to a study by Zábori et al. (2012), in which a major mode at around 180 nm was observed for RH smaller than 10%. A similar setup as in the recent study was used in the studies by Hultin et al. (2010, 2011) with similar results. In all the studies by Hultin et al. or Zabori et al. custom made DMPS systems built at ITM were used. However, this is equally true for the experiment by Mårtensson et al. (2003). It should also be noted that the differences between the mode diameter within the papers cited by the Referee (compare Mårtensson et al. (2003) with Keene et al. (2007) as plotted in Hultin et al. (2010)) are about as large as the difference between Keene et al. (2007) and for example Hultin et al. (2010). It is too early to yet conclude that the sea spray number mode is always centered at 100nm sharp. Since the sea spray studies in literature vary a lot (aerosol generation, water used, water/air flow rates, area available for bubble bursting) we unfortunately will not be able to explain the difference in the major sea spray modes between the different studies. The

authors agree that it is speculation that the peak at about 200 nm is caused by the jet system, but when no firm explanations can be offered, some degree of speculations should be allowed in a discussion section. Authors agree with Referee1 that the peak at about 200 nm is not explainable by the surfactants.
In Fig. 10 the two smallest OPC size bins measured are added (Dp 265 nm and Dp 290 nm) to the manuscript. In all other figures they are already included.

8. Revision:
The RH means during number size distribution measurements will be added to the figure captions.

The caption of Fig. 6 (p 19077) was changed to:
"Particle number size distributions from 35 g kg− 1 NaCl water for different water temperature ranges. Median size distributions were calculated for temperatures at 0 ◦C **(RH_{mean} = 29 %)**, between 1 ◦C and 4 ◦C **(RH_{mean} = 29 %)**, between 8 ◦C and 11 ◦C **(RH_{mean} = 30 %)** and between 13 ◦C and 16 ◦C **(RH_{mean} = 21 %)**. Dotted lines represent the 25th and 75th percentiles. Only every second data point is shown for clarity."

The caption of Fig. 7 (p 19078) was changed to:
"Normalized fitted particle number size distributions for different salinity ranges. **RH_{mean}= 43 % for salinities between 0 and 18 g kg^{-1}; RH_{mean}= 24 % for salinities between 18 and 35 g kg^{-1}"**.

The caption of Fig. 9 (p 19080) was changed to:
Particle number size distributions for different succinic acid concentrations **(RH_{mean} = 23 %)**. Water temperature medians are between 0.2 and − 0.2 ◦C and medians of salinity between 35.4 and 35.7 g kg^{-1}. Measurement time per concentration between 30 and 70 min. Grey shaded areas indicate the area between the 75th and 25th percentiles.

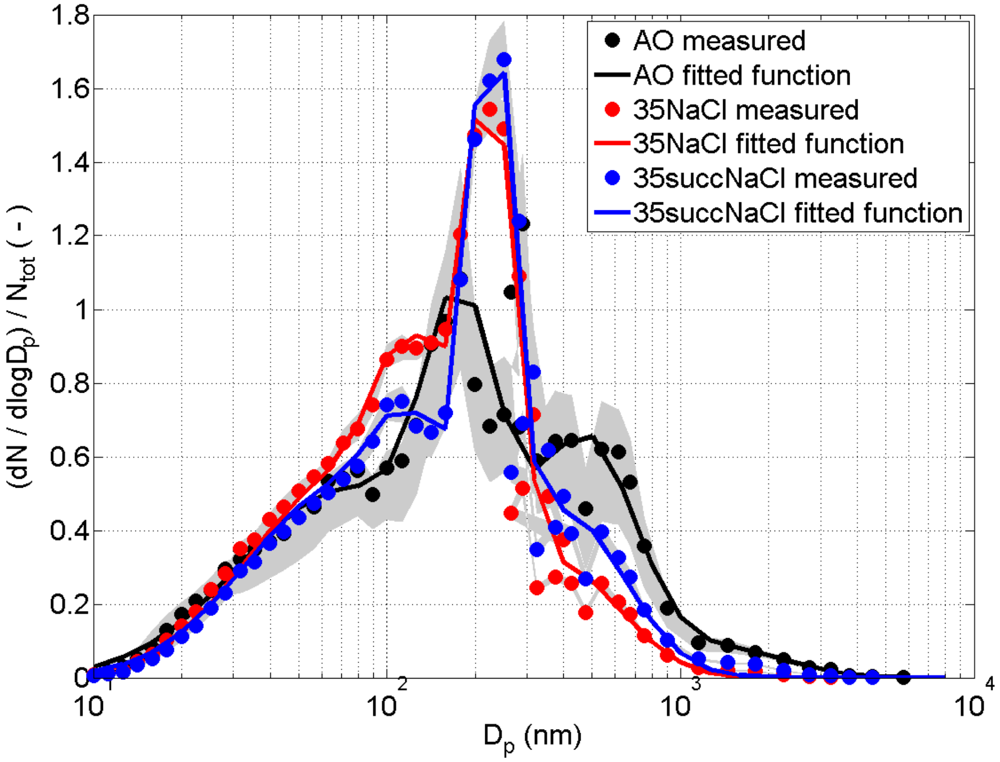
The caption of Fig. 10 (p 19081) was changed to:
Comparison between the normalized fitted particle number size distribution of Arctic Ocean water (water temperature median − 0.3 ◦C, salinity median 35.0 g kg− 1, **RH_{mean} = 7 %**), sodium chloride water without succinic acid (water temperature median 0.1 ◦C, median salinity 35.3 g kg^{-1}, **RH_{mean} = 29 %**) and sodium chloride water with succinic acid (water temperature median − 0.1 ◦C, median salinity 35.6 g kg− 1, **RH_{mean} = 23 %**). The normalized size distributions measured and the 25th and 75th percentiles (grey shaded areas) are shown in addition.

Since we cannot exclude the influence of RH on the particle size for the 0 – 18 g kg^{-1} salinity range in Fig. 7, the paragraph in line 22 p19051 – line5, p 19052 is changed from:
"To demonstrate the influence of salinity on particle number size distributions for the salinity range 0–35 g kg^{-1}, log-normally fitted size distributions normalized to the integral number density for the different salinity bins were compared (Fig. 7). Relative particle number concentrations for particles between 0.10 µm<D p <0.23 µm are higher for the lower salinities and decrease with increasing salinity until they remain stable at a salinity of about 15 g kg^{-1}. The dominant mode seen in Fig. 7 also has a slight shift from about 0.142 to 0.225 µm Dp when the salinity increases from 0–3 g kg^{-1} to 12–15 g kg^{-1}, above which the

mode diameter is stable. Relative particle number concentrations for smaller and larger sizes differ as well for different salinities, but not in an identifiable direction. One exception is an indication of a weak tail of smaller particles from about 15 g kg^{-1}. Generally, it can be said that there is a local maxima for all salinity bins at around 0.225 µm and the number size distributions do not vary considerably in relative shape for salinities higher than 15 g kg^{-1}."
To:
"To demonstrate the influence of salinity on particle number size distributions for the salinity range 0–35 g kg^{-1}, log-normally fitted size distributions normalized to the integral number density for the different salinity bins were compared (Fig. 7). Relative particle number concentrations for particles between 0.10 µm<D p <0.23 µm are higher for the lower salinities and decrease with increasing salinity until they remain stable at a salinity of about 15 g kg^{-1}. **As the relative humidity is rather high for the salinity range 0 -18 g kg^{-1}, it cannot be assumed that the particle diameters can be considered as dry diameters. Therefore, it might be that concentrations for real dry diameters are shifted towards smaller particle sizes. However, this would reveal the evolution observed in Fig. 7.** The dominant mode seen in Fig. 7 also has a slight shift from about 0.142 to 0.225 µm Dp when the salinity increases from 0–3 g kg^{-1} to 12–15 g kg^{-1}, above which the mode diameter is stable. Relative particle number concentrations for smaller and larger sizes differ as well for different salinities, but not in an identifiable direction. One exception is an indication of a weak tail of smaller particles from about 15 g kg^{-1}. Generally, it can be said that there is a local maxima for all salinity bins at around 0.225 µm and the number size distributions do not vary considerably in relative shape for salinities higher than 15 g kg^{-1}."

Additionally, the sentence: "However, since relative humidity was always lower than 45% during the experiments, all particle sizes are referred to as dry diameters" (line 21-22, p19042), is changed to:
"**The mean relative humidity (RH_{mean}) during all experiments was lower than 30 %, except for one case, where it was 43 %. Particle sizes measured at a mean relative humidity smaller than 30 % are considered to have dry diameters. This is not the case for the particle sizes with a mean relative humidity of 43 %. In the following text this will clearly be pointed out.** "

Figure 10 was modified like stated in Reply 10, the values of the two smallest size bins of the OPC were added (265 nm and 290 nm):



9. Comment:

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

9. Reply: Stirring was not needed to maintain a homogeneous salinity. Thank you for pointing out this sentence. When adding NaCl the water was stirred to dissolve the NaCl faster.

9. Revision:

The sentence: “Since the setup did not allow for maintaining cold temperatures and a homogeneous salinity at the same time (stirring the water with the bucket in the tank was very cumbersome), it was decided to conduct the experiment with water at room temperature.” (lines 21-24, p19045) was changed to:

“Since the setup did not allow for maintaining cold temperatures and **stirring the water to dissolve NaCl faster** at the same time (stirring the water with the bucket in the tank was very cumbersome), it was decided to conduct the experiment with water at room temperature.”

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

10. Reply:
In line 14, p 19048 it is written:
"In addition, measurements based on electrical mobility of the particles are not influenced by their color and likely less influenced by their shape compared to measurements based on optical properties." The sentence was thought as additional information about differences between DMA and OPC. We agree with the Referee that this sentence requires more detailed discussion. But we don´t think that this discussion about the differences in OPC and DMA regarding the influence of absorption and particle shape on the measured diameter, will support our choice to omit the first OPC bins:
First of all did we measure to large part water with added NaCl. In these cases, we can consider the influence of absorption as very small. Further on is the influence of the shape on the measured particle diameter likely insignificant, as the high numbers of particles cancel the effect out (due to the average of different shaped particles).
Therefore we deleted the sentence.

Please note that due to the deletion of the original Fig. 4 in the revised manuscript, the numeration of the figures changed.

Additional changes to the manuscript:
The sentence:"To demonstrate the influence of salinity […] log-normally fitted size distributions […] were compared (Fig.7)."(line 22-25) was changed to start at a new line.

References

Bauch, H. A. and Polyakova, Y. I. (2003): Diatom-inferred salinity records from the Arctic Siberian Margin: Implications for fluvial runoff patterns during the Holocene, Paleoceanography, 18, 1027.

Environmental Working Group (EWG) (1998): Oceanography atlas for the summer period, in Joint U.S.-Russian Atlas of the Arctic Ocean [CD-ROM], Univ. of Colo., Boulder, Colo..

Giles, K. A., Laxon, S. W., Ridout, A. L., Wingham, D. J., Bacon, S. (2012): Western Arctic Ocean freshwater storage increased by wind-driven spin-up of the Beaufort Gyre, Nature Geoscience, 5, 194-197.

Keene et al. (2007): Chemical and physical characteristics of nascent aerosols produced by bursting bubbles at a model air-sea interface, Journal of Geophysical Research, 112, D21202.

Macdonald, R. W., McLaughlin, F. A., Carmack, E. C. (2002): Fresh water and its sources during the SHEBA drift in the Canada basin of the Arctic Ocean, Deep-Sea Research I, 49, 1769-1785.

McPhee, M. G., Stanton, T. P., Morison, J. H., Martinson, D. G. (1998): Freshening of the upper ocean in the Arctic: Is perennial sea ice disappearing?, Geophysical Research Letters, 25, 1729-1732.

Nielsen, M. H., Erbs-Hansen, D. R., Knudsen, K. L. (2010): Water masses in Kangerlussuaq, alarge fjord in West Greenland: the processes of formation and the associated foraminiferal fauna, Polar Research, 29, 159-175.

Raskoff, K. A., Hopcroft, R. R., Kosobokova, K. N., Purcell, J. E., Youngbluth, M. (2010): Jellies under ice: ROV observations from the Arctic 2005 hidden ocean expedition, Deep-Sea Research II, 57, 111-126.

Stepanova, A., Taldenkova, E., Simstich, J., Bauch, H. A. (2007): Comparison study of the modern ostracod associations in the Kara and Laptev seas: Ecological aspects, Marine Micropaleontology, 63, 111-142.

Zábori, J., Krejci, R., Ekman, A. M. L., Mårtensson, E. M., Ström, J., de Leeuw, G, Nilsson, E. D. (2012): Wintertime Arctic Ocean sea water properties and primary marine aerosol concentrations, Atmospheric Chemistry and Physics Discussion, 12, 16085-16130.