Response to anonymous Referee#2: Zábori et al., 2013.

The authors thank anonymous referee#2 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 authors use two different aerosol particle sizing instruments, i.e. a DMPS for sub 0.3 micrometer particle sizes and an OPC for the range above. However those do not seem to have been matched. p. 31162, 2nd paragraph.: "The first two OPC bins were not used in the analysis of the winter data, and subsequent calculations, but are presented for completeness. There is evidence that for the overlapping size range of the DMPS and OPC instruments, the DMPS measurements provide higher quality data." Why is that? One questionable assumption for the particle instruments is the particle density, which requires a certain assumption for all particles, although each particle mass may display a different one. I am not totally sure that coarse particles display the same as submicron ones. The aspect of water content is as well critical. Was the water removed or is this a cocktail out of aerosol particle and water? Is there a method discriminating the water droplets from particles? Since both instruments use different inlet systems, dilutions and temperatures this might affect the overlapp, too.

1. Reply:
Tests with the GRIMM 1.109 have shown that its counting efficiency decreases considerably for particle diameters smaller than 300 nm and reaches a counting efficiency as low as about 15 % for the smallest diameter (265 nm) (Heim et al., 2008). This would imply for our results that the particle number concentration would be even higher with about 1000 particles cm^{-3} when assuming a counting efficiency of 50% for particles with a Dp of 265 nm. This more or less immediately increase of particle number concentration from one size bin to the next seems to be spurious, especially as this pattern is not observed for the overlapping DMPS measurements. The OPC is more sensitive to optical properties of the aerosols compared to the DMA. The RH was less than 30% in the sampling inlet used by both instruments. If the reviewer by particle density referees to optical properties, then the reviewer has a point that a change in chemical proprieties around the overlapping range could result in spurious results. Unfortunately we do not have that detailed chemical information. We cannot explain the “outliers” in the OPC. However, they do not influence the conclusions drawn.

2. Comment:
The focus has been set to produced aerosol number concentrations, but the information on volume or mass is missing. This is questionable, since aerosol dynamics might have transfered the particles formed to larger sizes, i.e. the same amount of water soluble material but a smaller amount of particles formed. The aerosol dynamics has been removed in a variety of scaling and normalizing steps, which are certainly helpful to make the big variety of size distributions comparable. Nevertheless this point might impact on the very hypothetic conclusions drawn.

2. Reply:
In Zábori et al. (2012a) particle number volume size distributions were calculated for the lower water temperature ranges (from -2 to 5 °C) for which a decrease in particle number concentration was observed. It was shown that the volume size distributions followed the particle number size distribution behavior (smaller volumes for higher water temperatures) and it can be concluded, that the trend of decreasing particle numbers with an increase in water temperature for the lower water temperature range is not dominated by particle dynamics, but by particle emissions. In the revision of the manuscript this will be stressed. However, to further enhance the discussion in the manuscript, particle volume size distributions were calculated for data presented in Fig. 4-6. The particle volume size distributions follow the pattern of the number size distributions, even though the difference in volume for particles with a Dp < 200nm is hardly visible due to the large size range covered and the volume dependence on the 3rd power of the diameter.

2. Revision:
To stress that the particle number concentration decrease with an increase in water temperature at the lower water temperatures is a consequence of the emissions and not of particle dynamics, the revised manuscript refers to the study by Zábori et al. (2012a). In section 3.1 on page 31163 after line 9 the following sentences were added:

"**The observed decrease of particle number concentrations with increasing water temperatures up to about 5 – 7 °C (cf. Fig. 3 a-c) is a result of a decrease in particle emissions rather than a result of particle dynamics as shown in Zábori et al. (2012a).**"

Results of the calculated volume size distributions from the data presented in Fig. 5-7 are added after line 9 on page 31166:
"**Median volume size distributions for the data presented in Figs. 4–6 are shown in Figs. 7–9. The maxima of the volume distribution occur at dry diameters between 3 and 4 μm for the winter measurements and between 2 and 3 μm for the summer measurements. The volume size distribution changes in a similar manner to the particle number size distribution, even though the differences between winter and summer are more distinct for the larger particles when comparing volume due to the size dependence of the volume calculations.**"

Particle volume size distributions for the data presented in Fig. 4-6 were added to the manuscript and named Fig. 7-9. Consequently, Fig.7-9 is renamed to Fig. 10-12.The reference to the figures in the text was changed accordingly to the new figure names.

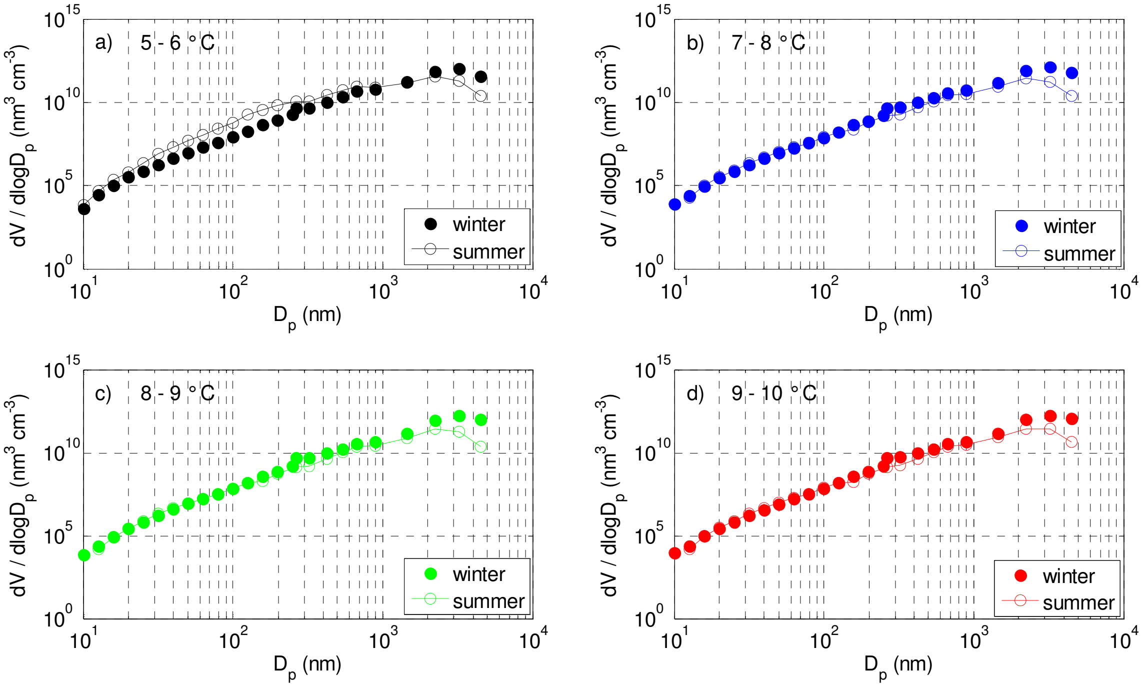

Fig. 7. Comparison of particle volume size distributions resulting from water sampled close to the glacier during summertime and wintertime, for the same water temperature ranges of (a) 5–6 °C (b) 7–8 °C (c) 8–9 °C (d) 9–10 °C. For clarity only every second data point is shown.

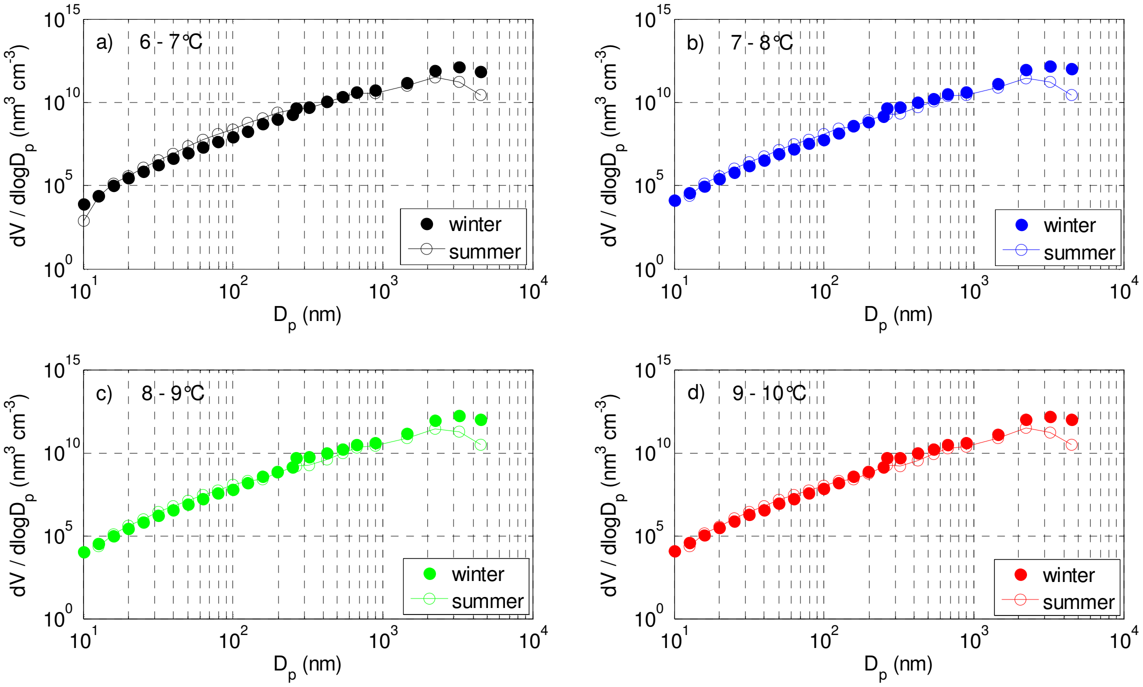

Fig. 8. Comparison of particle volume size distributions resulting from water sampled close to the fjord mouth during summertime and wintertime, for the same water temperature ranges of (a) 6–7 °C (b) 7–8 °C (c) 8–9 °C (d) 9–10 °C. For clarity only every second data point is shown.

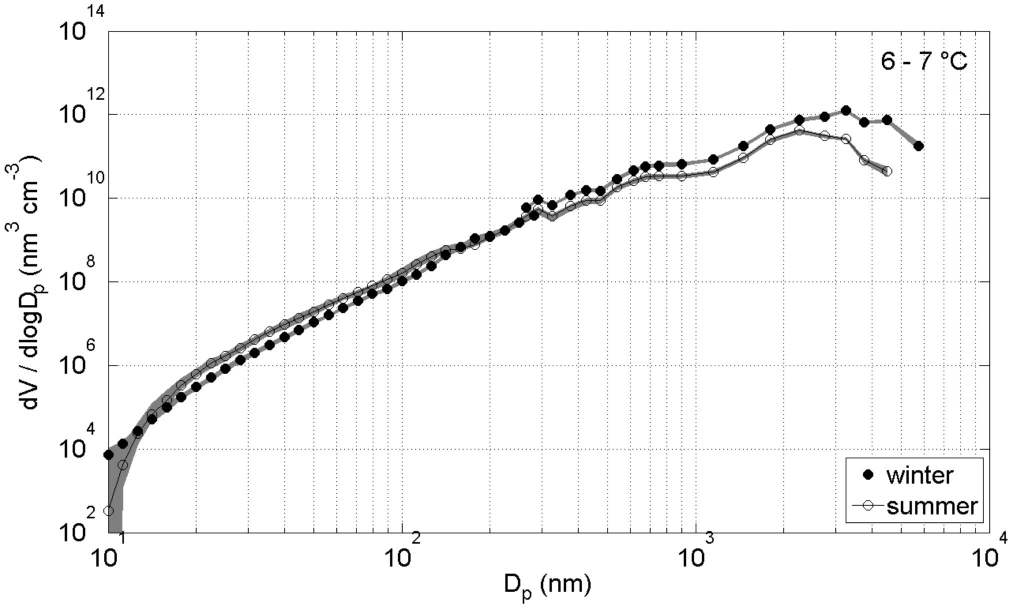

Fig. 9. Comparison of particle volume size distributions resulting from deep sea water during summertime and wintertime, for the same water temperature ranges of 6–7 °C. Grey shaded areas represent the interquartile ranges.

3. Comment:
Have the authors tried Gaussian fits to the size distributions measured? This would provide additional information to the median diameter, which is certainly valuable information but the mode width might provide additional information.

3. Reply:
Gaussian distributions were not fitted to the data, but based on the comment, lognormal functions for median winter and summer number size distributions (Fig4 and Fig5) were fitted and the parameters were added into a table. For the respective medians, the lowest water temperature range (5 – 6 degC,close to glacier; 6 - 7 degC close to fjord mouth) were not considered (please see lines 18-25, page 31167 for explanation).

3. Revision:

Table 2: Parameters for fitted lognormal aerosol number size distributions, where Nx is the number concentration (cm^{-3}), Dgx is the geometric diameter (µm) and σx is the standard deviation for the different modes and seasons.

	winter	**summer**
N1	**129**	**182**
Dg1	**0.063**	**0.067**
σ1	**2.48**	**2.48**
N2	**22**	**28**
Dg2	**0.164**	**0.227**
σ2	**1.19**	**1.08**
N3	**132**	**58**
Dg3	**0.43**	**0.575**
σ3	**1.54**	**1.50**
N4	**58**	**18**
Dg4	**2.1**	**1.8**
σ4	**1.42**	**1.34**

The table and the following text is inserted after line 9 on page 31166: "**In order to characterize the particle number size distributions, lognormal functions were fitted to the median particle number size distributions of the summer and winter data presented in Fig. 4 and Fig. 5. When calculating the medians for the respective season, the lowest water temperature range for the water sampled close to the glacier and the water sampled close to the fjord mouth were not considered (cf. lines 18-25, page 31167). The parameters of the fitted lognormal distributions are presented in Table 2.**

4. Comment:
Different flows and heights of inflow during summer- and in wintertime: What about the influence of this on the results? Are there any tests about this aspect?

4. Reply:
Unfortunately, no systematic tests with different pump heights and consequently water flow rates have been conducted. However, as stated in the manuscript (cf., lines 9-11, page 31167) some data from the winter experiments were available to check the influence of two

different pump rates on the shape of the size distribution. Nevertheless, the difference is not exactly the same as the difference between the winter and summer experiments, and we cannot completely exclude that the shape was not influenced by the different water flow rates.

5. Comment:
Figure 3: Two points: (1) Why is there an "outlier" for 6 degree Celsius in summertime (plots b and c)? Have the datasets been checked for outliers? Since commonly multiple charge corrections are applied this feeds back into the results of other sections.
(2) I can see a decoupling of larger and smaller particle numbers during summer with no clear temperature dependency during (subplot d). Perhaps this results of the inflow of glacier water, vertical mixing inside the Kungsfjorden or an additional source during summertime for smaller particles. Actually the results stated in the text (p. 31163, top paragraph) should be provided with standard deviations. Checking the summertime values it seems both values could match still within their uncertainty range.

5. Reply:
(1) The data shown are medians of particle number concentrations for the certain water temperature ranges (and the different experiments), so the mean is quite robust against outliers. We are not sure why the concentrations around 6 degC are out of the line, but we can speculate that the setup was not yet stable (cf. lines 18-23, page 31167).

(2) We thank the reviewer for the suggestions. In the revised manuscript a paragraph will be added where Fig. 3d is discussed. In addition the uncertainties asked for were added.

5. Revision:
(2) The sentence "However, it should be noted that the interquartile range is relatively large (Fig. 3b, c)." (lines 8-9, page31163) is replaced by the sentences: "**The uncertainty range (between the 75th percentiles and 25th percentiles of the data) of particle number concentrations for water sampled close to the glacier ranges between 1593 and 622 particles cm^{-3} during summertime and between 662 and 417 particles cm^{-3} during wintertime for a median water temperature between 5 and 6°C (cf., Fig. 3b). The same uncertainty range of particle number concentrations of water sampled at the fjord mouth ranges between 1119 and 675 particles cm^{-3} for water sampled during summertime and between 661 and 521 particles cm^{-3} for water sampled during wintertime for median water temperatures between 5 and 6°C (cf., Fig. 3c).**"

In the discussion chapter the uncertainty is added as well. The sentence: "The relatively large interquartile range observed for the summertime lowest Tw range also supports the conclusion that the system was unstable for these measurements." (lines 23-25, page 31167) was modified to "**The relatively large interquartile range between the 75th and 25th percentiles (971 particles cm^{-3} and 444 particles cm^{-3} for the water sampled close to the glacier and at the fjord mouth, respectively) observed for the summertime lowest Tw range also supports the conclusion that the system was unstable for these measurements.**"

Some discussion about Fig. 3d is added after line 29 on page 31167:

“**Figure 3d shows that the ratio between particles with a Dp > 0:01µm and with a Dp > 0:250µm is a function of water temperature during wintertime but not during summertime. The ratio is also on average higher for summertime measurements compared to wintertime measurements. This results from a combination of both higher particle number concentrations for particles with a Dp < 250 nm and lower particle number concentrations for particles with a Dp > 250 nm during summertime. There are a number of possible reasons for this. Firstly, the water composition is different due to high biological activity during the summer season compared to the winter season. Secondly, the local sources of melt water and river runoff in the fjord change with season which possibly modified the measured particle concentrations at different sizes. Thirdly, a potentially important factor for substances present on the ocean surface is the vertical mixing of the water column, bringing material from different depths to the surface. Which, if any, of these factors, or combination of factors, led to the differences in the ratios between the measured particles with a Dp >0:01µm and with a Dp >0:250µm between summer- and wintertime measurements is impossible to say with the data available from this study.”**

6. Comment:
As pointed out by a different reviewer there are some curiosities among the size distributions shown: (a) In Fig. 4 each winter particle size distribution shows an "outlier" at around 280 nm in diameter. This is strange and would point on a certain primary particle source of a distinct size especially if it is being observed for four different experiments. (b) It comes even more strange in this figure to see an identical particle size distribution for all four cases in wintertime (hypothesis of mine) with every small up and down reproduced in every of the subplots. The feature is a little bit hidden in subplot a) as the summertime values display a higher magnitude. Has one mean measurement been used for all of the intercomparisons? This would explain parts of the results and appears even more pronounced in Fig. 5.

6. Reply:
a) please see reply to the 1st comment.
b) We thank the reviewer for the comment. The presented winter and summer data in Fig. 4+5 are medians of the different water temperature ranges, meaning not one mean measurement has been used for all of the intercomparisons. However, based on the comment, the data has been reanalyzed and it has been recognized that for the water sampled close to the fjord mouth (Fig. 5) the median of the DMPS measurement for the temperature range 7 to 8 degC (wintertime) was used for the temperature range 9 to 10 degC as well. This was corrected and the data shown in Fig.8 were recalculated for the temperature range 9 to 10 degC, as well. Due to the corrections the relative deviation between summer and winter data for Dp < 0.125 µm changed from about 60% to about 46%. The deviation for particles with Dp > 125 µm when comparing summer and winter data did not change (line 29 on the page 31165 to line 2 on the page 31166).
All in all, the size distribution shapes and magnitudes are very similar for the presented water temperature ranges which can be addressed by the insensitivity of the particle number concentrations when water temperature changes occur above about 6 degC.

6. Revision:

Based on the reply Fig. 5 and Fig. 8 were corrected:

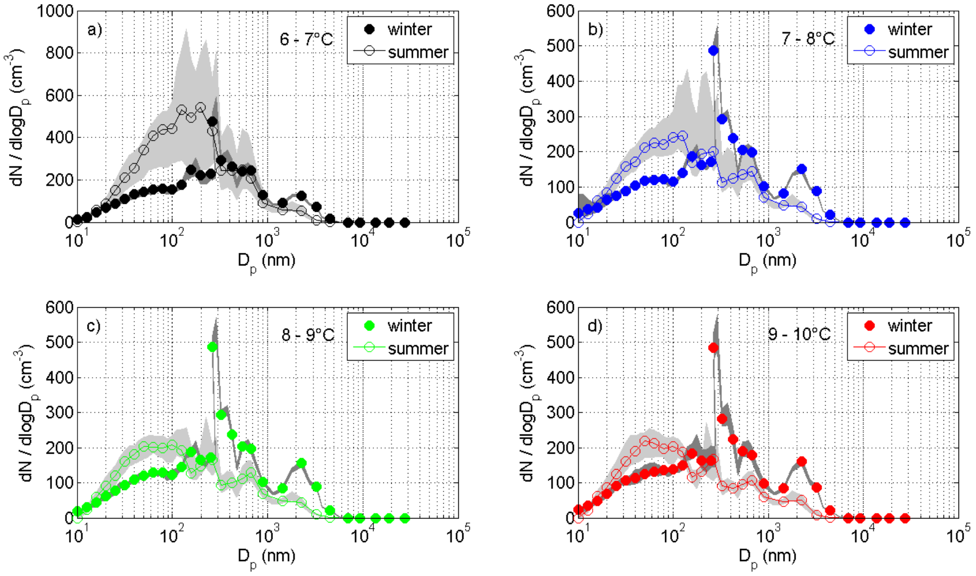

Fig. 5. Comparison of particle number size distributions resulting from water sampled close to the fjord mouth during summertime and wintertime, for the same water temperature ranges of (a) 6–7 °C (b) 7–8 °C (c) 8–9 °C (d) 9–10 °C. For clarity only every second data point is shown. Grey shaded areas represent the interquartile ranges. Note the different scale for the lowest temperature range.

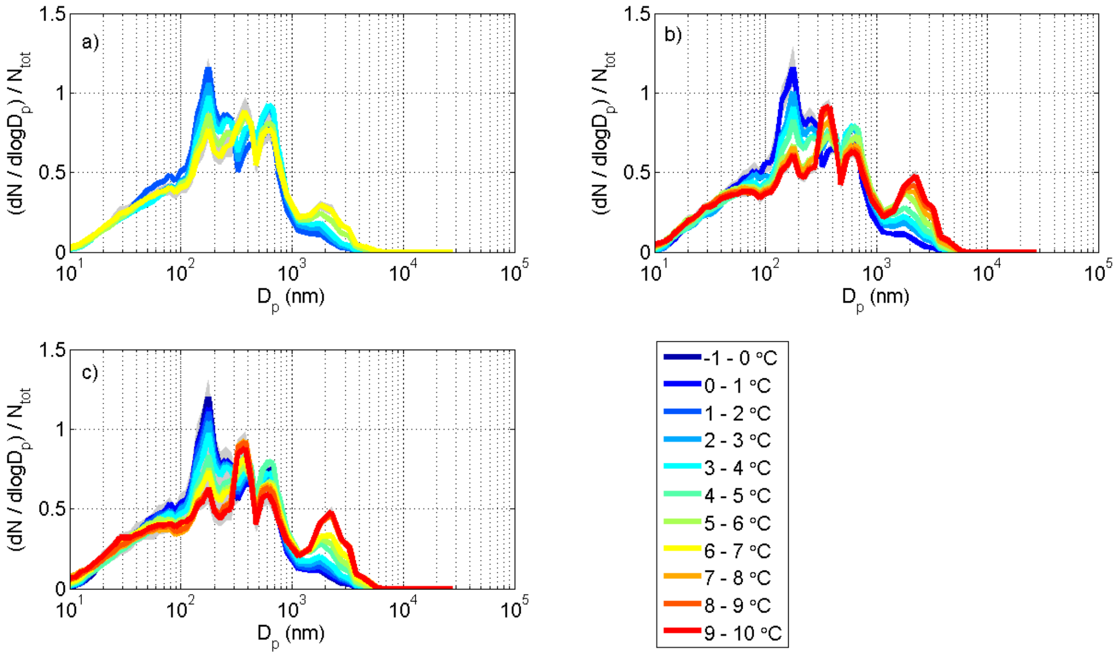

Fig. 8. Normalized number size distributions for winter experiments resulting from bubble bursting of (a) deep fjord water (b) water sampled close to the glacier (c) water sampled at the fjord mouth. Grey shaded areas represent the interquartile range for the lowest and highest water temperature range. The same grey shade is used for both temperature ranges and indicates if there is an obvious difference between the particle number concentrations of different sizes of the lowest and highest water temperature.

Due to the corrections the relative deviation between summer and winter data for Dp < 0.125 µm changed from about 60% to about 46%. The deviation for particles with Dp > 125 µm when comparing summer and winter data did not change. The sentence "Particle number concentrations are for Dp < 0.125µm about 60 % higher for summer than winter measurements, and for Dp > 0.125µm about 45 % lower for summer compared to winter." (line 29 on the page 31165 to line 2 on the page 31166), was changed to "**Particle number concentrations are about 46 % higher for Dp < 0.125µm during summer than winter measurements, and for Dp > 0.125µm about 45 % lower for summer compared to winter measurements.**"

7. Comment:
Fig. 6: What is the way of errorbar calculation, i.e. standard deviation or counting error? This applies in detail for the DMPS errorbars beacuse of the small charged fraction of particles.

7. Reply:
All presented data in the figures are medians and the associated uncertainties are presented by the area enclosed by the 25th and 75th percentiles. Based on the comment the counting errors were calculated for the data presented in Fig.6. It was with about 13 % highest for the particles with a Dp of 10 nm during summertime experiments (the counting error was about

8% and 6% for the particles with a Dp of 4.5 µm and 3.75 µm, respectively). For the wintertime experiments the error was smaller than 1% for all sizes.

The reply and revision of the manuscript based on the following comments 8 to 10 will be combined, as all the comments require a revision of the “Future impact” chapter in the manuscript. Moreover is the revision combined with changes made based on comments by reviewer#1.

8. Comment:
Figs. 7 and 8: It is a good idea to plot the particle size distributions for different water temperatures of the original samples. The striking difference between the behaviour during winter and summertime is worth more discussion with respect to the implications for the presented hypothesis. One would expect similar behaviour.

9. Comment:
This indicates that the total feedback loops are more complex than captured so far by a simple temperature based particle production. The authors really discuss their results in a good way. However the hypothesis is still fragile. Certainly I agree with the authors that more research is needed and that this paper presents a hypothesis to work on, but the present data delivers only indications but no final proof, since studies have been conducted in an arbitrary environment (PET volume) with filtered air.

10. Comment:
What happens if there is an additional feedback loop including rising atmospheric background particle numbers with temperature that originate elsewhere and which are transported towards the site of interest? Their prospective lifetime would be sufficiently long. What about the size of air bubbles with rising temperatures? Size the solubility changes with temperature, those may become larger and compensate partially the observations being made. What about the surface wind speed initiating the wave breaking and the sea salt particle production (Table 1)? Does this force more intense vertical mixing during winter or what about its impact on the conclusions drawn?

11. Comment:
I evidently miss the inclusion of the chemical informations of the former article of the group (Zabori et al., 2012) that provides salinity, dissolved organic compounds etc. Do these parameters change with sea surface temperature, capable in explaining the observations? Therefore I’d like to see a broader discussion of potential effects if such a hypothesis is being concluded.

Based on all of the above comments and comments by reviewer 1, the section “Future implication” has been revised completely.
Based on comments of reviewer 1 Fig.9 (new Fig. 12) was changed to:

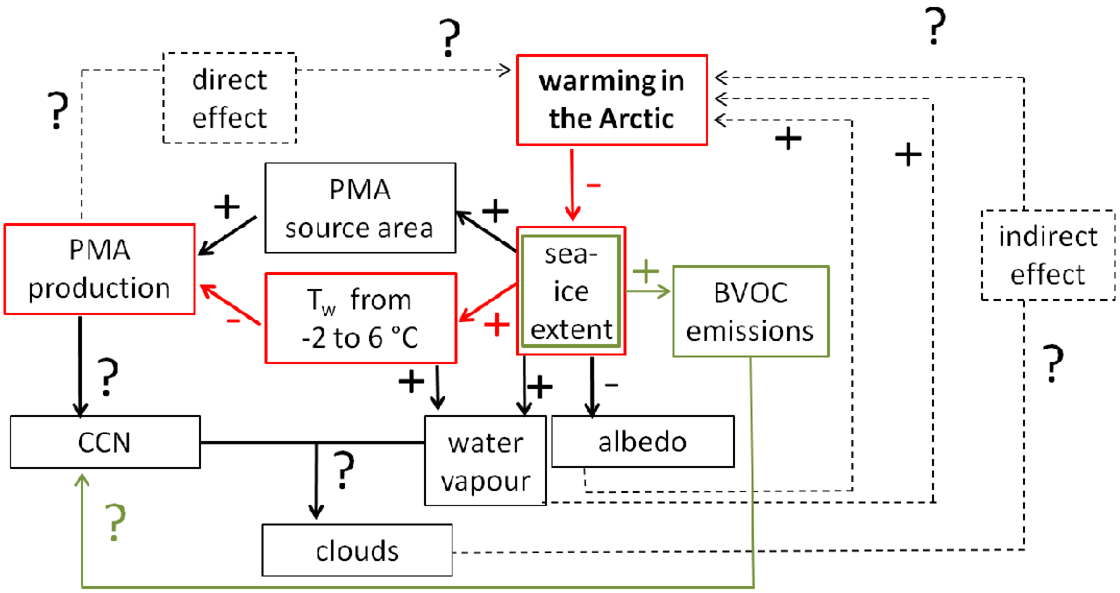

Fig. 12. Potential feedback loop resulting from a warming in the Arctic. Plus signs indicate increases and minus signs indicate decreases. Question marks indicate that the direction of the change is not clear. **The focus of this study is marked by the red colour. The green colour indicates the path related to changes in BVOC emissions.**

5 Future implications

The decrease in Arctic sea ice extent due to climate warming, will be followed by changes in a number of different processes, which eventually will lead to different feedbacks. Figure 12 displays a potential feedback loop focused on processes closely related to PMA production and changes in sea ice cover (marked in red). For the sake of completeness we have also included one BVOC emission process loop (biogenic volatile organic compounds; which also include DMS) in the figure (marked in green). It is worth reiterating that our results are based on laboratory experiments and therefore lack the interdependencies that may occur in the Arctic environment. However, to our knowledge no existing in-situ measurements have been reported in the literature. As such we feel it appropriate to base our discussion on possible Arctic feedbacks on our laboratory measurements. With this caveat in mind it should be noted that Fig. 12 only serves to exemplify key pathways and does not represent all possible feedback processes related to aerosol direct and indirect effects that may be affected by climate change in the region. Following discussion of the feedback loop, analysis of those parameters which are expected to alter with climate change is conducted. However, these parameters are not included in the figure to retain clarity. The decrease in Arctic sea-ice extent, which follows from a warming of the Arctic, will influence parameters controlling the production of primary and secondary marine aerosols (Fig. 12). These factors are the increase of the PMA source region, an increase in sea surface temperature (with subsequent effects on PMA production as presented within this study), and an increase in BVOC emissions, those precursor gases responsible for secondary marine aerosol production.

If sea surface temperature changes in the region Tw >~6°C, no sea ice will be present and no change of PMA due to a change in source area will occur. If the results of our laboratory

studies are correct there will also be little effect of sea surface temperature changes on PMA production at these temperatures as effects here are minimal (cf., Sect. 3.1). Assuming a change of Tw in the lower water temperature range (Tw < ~6°C), the net resulting change in the PMA production due to the two factors (increasing Tw and increasing source area) is currently unknown (due to their opposing sign on PMA production). This issue therefore merits further studies, for example using modeling tools, as well as including additional parameters other than Tw that may impact on PMA production. It is likely that there will be a region that experiences sea surface temperature changes in the range between seawater ice formation and Tw<~6 °C. In a scenario with no or rather limited sea ice extent PMA production will mainly be affected by the temperature trend.

The change in PMA production has important implications for the number of available cloud condensation nuclei (CCN) in the Arctic region. An increase in water temperature below ~6 °C will likely decrease the PMA number concentration, and subsequently the amount of potential CCN (cf., Zábori et al., 2012a). Further to this, our results highlight that the temperature dependent relative PMA number size distribution is changing in the same direction for the accumulation mode but in different directions for the Aitken and coarse mode for aerosols produced by water sampled during wintertime and summertime. An increase in water temperature (from 5 to 13 °C for summer experiments and from -1 to 9 °C for winter experiments) reduces the relative particle number concentration in the accumulation mode i.e. those particles that are most efficient as CCN. The different changes for the Aitken and coarse mode for the different seasons may affect the particle dynamics and consequently the amount of CCN which can be activated into cloud droplets. Besides primary marine aerosols, the importance of secondary marine aerosols as CCN is currently being discussed extensively in the literature. Conclusions on the role of DMS for CCN production are ambiguous. Charlson et al. (1987) showed that the CCN number was affected by DMS emissions however, other studies have found only sporadic or no correlation between DMS and CCN (e.g. Bates et al., 1992 and Berresheim et al., 1993 in Kloster et al.,2006). Gabric et al. (2005) modeled the DMS production in the Arctic Ocean as a result of a CO_2 tripling up to the year 2080. DMS emissions were shown to increase considerably, mainly due to the decrease of the sea-ice extent and the resulting decrease in the ocean-atmosphere exchange barrier. Besides DMS, other BVOCs are suspected to be precursors of secondary aerosols, e.g. isoprene (Vaattovaara et al., 2006). A study modeling the Arctic Ocean has highlighted the importance of both DMS and other BVOCs for new particle formation (Karl et al., 2012). Despite these hints it is our opinion that the factors influencing BVOC emissions in the Arctic Ocean are not well enough constrained to discuss the potential influence of climate change (see Shaw et al., 2010 for a review of marine isoprene and monoterpene emissions).

Our limited understanding of Arctic CCN characteristics and their potential response to a changing climate affects our ability to predict future aerosol related changes in cloud cover. The supersaturation of water vapour in the atmosphere, atmospheric dynamics and the availability, size, and composition of CCN determines if and where cloud droplets and clouds form. The amount of water vapour in the Arctic atmosphere is dependent on sea ice cover, sea surface temperature and air temperature. Lower sea ice cover coincident with an increase in sea surface temperature should result in an increased transport of water vapour to the atmosphere given that the sea ice represents an evaporation barrier for the water and higher sea surface temperatures will increase evaporation rates. As the air

temperature increases more water vapour can remain in the atmosphere before condensation occurs consequently intensifying Arctic warming.

A further parameter besides PMA source area, sea surface temperature, BVOC emissions, and water vapour that is influenced by a decreasing sea ice extent is the surface albedo, which decreases with decreasing sea ice cover. This decrease in surface albedo, coincident with increases in water vapour, an unknown change in PMA production and the subsequent unknown change in cloud properties close the feedback loop and impact on the warming of the Arctic. Both the decrease in surface albedo and the increase in water vapour are expected to have a positive feedback on the warming of the Arctic.

It is impossible to assess the net sign and magnitude for all the feedbacks in the warming Arctic linked to changes of sea ice coverage and PMA production at this point. Nevertheless we can speculate that the non-linear relation between PMA production and increasing sea surface temperatures in concert with changes in sea ice cover have a strong potential to contribute to the evolution of the Arctic climate and deserve future attention.

As previously mentioned, the feedback loop presented in Fig. 12 only considers a small fraction of those factors important for marine aerosol production in the Arctic. What follows is a discussion of other parameters that may impact on PMA production in the Arctic Ocean.

The salinity of the Arctic Ocean is expected to decrease in future for a number of different reasons; at local scales a negative mass balance of glaciers has been observed (Nuth et al., 2010). This results in a decrease in the salinity of ocean surface water. It has also been suggested that an interplay between wind fields, melting sea ice and river runoff may lower salinities in the Arctic Ocean (Giles et al., 2012; MacDonald et al., 2002). The effect of salinity on PMA is unclear. Zábori et al. (2012a) did not observe a clear trend of particle number concentrations with salinity changes between 26 and 36‰ when conducting experiments with Arctic Ocean water. However, a number of other studies have shown that higher salinities can result in higher particle number concentrations (Mårtensson et al., 2003; Tyree et al., 2007; Hultin et al., 2011). Nevertheless these case experiments were conducted outside the Arctic.

An increase in water temperature affects the production of gas bubbles in the water. These gas bubbles produce aerosols when rising and bursting on the ocean surface. Thorpe et al. (1992) concluded that the temperature related decrease in gas solubility and the temperature related increase in molecular diffusivity cancel out and that the net effect of a temperature increase is a decrease in bubble concentrations due to a reduction in viscosity. However, a water temperature independent change in oxygen saturations in the water occurs due to a change in photosynthesis rates. Since altered environmental conditions caused by climate change may impact the flora and fauna in the Arctic (Wassmann and Reigstad, 2011; Tremblay et al., 2011), a future change in oxygen saturation is likely to take place. Zábori et al. (2012a) did not observe a change in PMA number production with a change in oxygen saturation between 72 and 83% for Arctic Ocean conditions. Hultin et al. (2011) observed for Baltic Sea water an anti-correlation between particle production and oxygen saturations in the range 90 to 100 %. Thus, it is not possible to derive a clear conclusion on the role of oxygen saturation on PMA production.

The concentration of organic substances in the water is known to impact PMA production. Depending on the substance used, differences in total particle number concentrations and shifts in particle number size distributions have been observed. Experiments with

dissolved organic carbon and colloidal organic matter (exudates of algae) have produced a shift of the size distribution towards smaller particle sizes relative to artificial inorganic seawater (Fuentes et al., 2010b). Tyree et al. (2007) observed an increase in total particle number concentration with an increase in the concentration of oleic acid. Zábori et al. (2012b) observed a decrease in total particle number concentration with an increase in the concentration of succinic acid. Clearly the influence of organic substances on PMA is complex and there is a need for further well designed experiments to aid understanding.

Another important parameter for PMA production that is impossible to assess in laboratory experiments using water jets is the wind speed. It has been shown that PMA production is a non-linear function of wind speed (Lovett, 1978; Nilsson et al., 2001). Struthers et al. (2011) simulated the sea salt emissions in a future Arctic climate, where the emissions were dependent on sea ice extent, sea surface temperature and the 10m wind speed. An increase in sea salt emissions was observed with the effect driven predominantly by the decrease in sea ice extent and changes in sea surface temperature rather than from a change in wind speed. However, a change in wind speed is likely to change the organic composition of PMA. Gantt et al. (2011) developed a conceptual model describing the organic mass fraction of sea spray aerosols as a function of wind speed. As the wind speed exceeds 3-4 ms^{-1} the surface microlayer (a tens of nanometers to about 100 µm thick layer on the ocean surface consisting of organic substances (Lion and Leckie, 1981; Cunliffe et al., 2013) starts to get mixed with subsurface water and at wind speeds larger than 8 ms^{-1} a homogeneous water column is expected, resulting in lower organic mass fractions of sea spray aerosols. Transferred to our experiments, this means that possible PMA organic fractions in the aerosols are not only dependent on the production of the organic substances in water, but also on the mixing in the water column.

Additional factors potentially important for the climate feedback loop shown in Fig. 12 are an increase in the maritime transportation, tourism, and oil exploration activity as a result of an increasingly ice-free Arctic Ocean. These will result in increasing anthropogenic aerosol emissions and the release of substances into the Arctic Ocean water.

Our attempt is to present a rather simplified scheme of possible feedbacks linked to PMA production from the Arctic Ocean and sea water properties. There are numerous other processes influencing the ocean-atmosphere interactions. Some of them were discussed in the text and some of them not at all, like impact of broader scale processes outside the Arctic linked to the atmospheric and ocean general circulation.

References:

Bates, T. S., Calhoun, J. A., and Quinn, P. K. (1992): Variations in the methanesulfonate to sulfate molar ratio in submicrometer marine aerosol-particles over the South-Pacific ocean, J. Geophys. Res., 97, 9859–9865.

Berresheim, H., Eisele, F. L., Tanner, D. J., McInnes, L. M., Ramseybell, D. C., and Covert, D. S. (1993): Atmospheric sulfur chemistry and Cloud Condensation Nuclei (CCN) concentrations over the northeastern Pacific coast, J. Geophys. Res., 98, 12 701–12 711.

Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G. (1987): Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, Nature, 326, 655 – 661.

Fuentes, E., Coe, H., Green, D., de Leeuw, G., and McFiggans, G. (2010): On the impacts of phytoplankton-derived organic matter on the properties of the primary marine aerosol -- Part 1: Source fluxes, Atmos. Chem. Phys., 10, 9295—9317.

Gabric, A. J., Qu, B., Matrai, P., and Hirst, A. C. (2005): The simulated response of dimethylsulfide production in the Arctic Ocean to global warming, Tellus, 57B, 391–403.

Gantt, B., Meskhidze, N., Facchini, M. C., Rinaldi, M., Ceburnis, D., and O'Dowd, C. D. (2011): Wind speed dependent size-resolved parameterization for the organic mass fraction of sea spray aerosol, Atmos. Chem. Phys., 11, 8777-8790.

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

Heim, M., Mullins, B. J., Umhauer, H., and Kasper, G. (2008): Performance evaluation of three optical particle counters with an efficient "multimodal" calibration method, J. Aerosol Sci., 39, 1019-1031.

Hultin, K. A. H., Krejci, R., Pinhassi, J., Gomez-Consarnau, L., Mårtensson, E. M., Hagström, Å., and Nilsson, E. D. (2011): Aerosol and bacterial emissions from Baltic Seawater, Atmos. Res., 99, 1–14.

Karl, M., Leck, C., Gross, A., and Pirjola, L. (2012): A study of new particle formation in marine boundary layer over the central Arctic ocean using a flexible multicomponent aerosol dynamic model, Tellus B, 64, 1-24.

Kloster, S., Feichter, J., Maier-Reimer, E., Six, K. D., Stier, P., and Wetzel, P. (2006): DMS cycle in the marine ocean-atmosphere system – a global model study, Biogeosciences, 3, 29–51.

Lion, L. W., and Leckie, J. O. (1981): The biogeochemistry of the air-sea interface, Annu. Rev. Earth Pl. Sc., 9, 449–486.

Lovett, R. F. (1978): Quantitative measurement of airborne sea-salt in the North Atlantic, Tellus. 30, 358–364.

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

Mårtensson, E. M., Nilsson, E. D., de Leeuw, G., Cohen, L. H., and Hansson, H.-C. (2003): Laboratory simulations and parameterization of the primary marine aerosol production, J. Geophys. Res.-Atmos., 108, 4297, doi:10.1029/2002JD002263.

Nilsson, E. D., Rannik, U., Swietlicki, E., Leck, C., Aalto, P. P., Zhou, J., and Norman, M. (2001): Turbulent aerosol fluxes over the Arctic Ocean 2. Wind-driven sources from the sea, J. Geophys. Res.-Atmos., 106, 32139—32154.

Nuth, C., Moholdt, G., Kohler, J., Hagen, J. O., and Kääb, A. (2010): Svalbard glacier elevation changes and contribution to sea level rise, J. Geophys. Res.-Earth Surf., 115, doi:10.1029/2008JF001223.

Struthers, H., Ekman, A. M. L., Glantz, P., Iversen, T., Kirkevåg, A., Mårtensson, E. M., Seland, Ø., and Nilsson, E. D. (2011): The effect of sea ice loss on sea salt aerosol concentrations and the radiative balance in the Arctic, Atmos. Chem. Phys., 11, 3459—3477.

Thorpe, S. A., Bowyer, P., and Woolf, D. K. (1992): Some factors affecting the size distributions of oceanic bubbles, J. Phys. Oceanogr., 22, 382—389.

Tremblay, J.- É., Bélanger, S., Barber, D. G., Asplin, M., Martin, J., Darnis, G., Fortier, L., Gratton, Y., Link, H., Archambault, P., Sallon, A., Michel, C., Williams, W. J., Philippe, B., and Gosselin, M. (2011): Climate forcing multiplies biological productivity in the coastal Arctic Ocean, Geophys. Res. Lett., 38, L18604.

Tyree, C. A., Hellion, V. M., Alexandrova, O. A., and Allen, J. O. (2007): Foam droplets generated from natural and artificial seawaters, J. Geophys. Res.-Atmos., 112, D12204.
Wassmann, P., and Reigstad, M. (2011): Future Arctic Ocean seasonal ice zones and implications for pelagic-benthic coupling, Oceanography, 24, 220–231.

Vaattovaara, P., Huttunen, P. E., Yoon, Y. J., Joutsensaari, J., Lehtinen, K. E. J., O'Dowd, C. D., 15, and Laaksonen, A. (2006): The composition of nucleation and Aitken modes particles during coastal nucleation events: evidence for marine secondary organic contribution, Atmos. Chem. Phys., 6, 4601–4616.

Zábori, J., Krejci, R., Ekman, A. M. L., Mårtensson, E. M., Ström, J., de Leeuw, G., and Nilsson, E. D. (2012a): Wintertime Arctic Ocean sea water properties and primary marine aerosol concentrations, Atmos. Chem. Phys., 12, 10405–10421.

Zábori, J., Matisans, M., Krejci, M., Nilsson, E. D., and Ström, J. (2012b): Artificial primary marine aerosol production: a laboratory study with varying water temperature, salinity, and succinic acid concentration, Atmos. Chem. Phys., 12, 10709–10724.