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

The authors thank anonymous Referee#2 for insightful comments on the manuscript. The reviewer provided several 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.

Major comments

1. Comment:
a) *It is arguable that succinic acid can be used as a proxy to represent the average behavior of marine biogenic organics. While marine organics have been shown to present an average molar mass of the order of 2 kDa (Moore et al., Fuentes et al.), succinic acid is a low molar mass compound that might not represent the behavior of organic biopolymer mixtures. Given its molecular size, succinic acid would only be representative of the seawater DOC fraction and not of the biogenic nanogels released by marine biota. The surface activity and the conformational changes of surfactants as a result of temperature and ionic concentration variations are highly dependent on the compound chain structure and on the interaction between the large biopolymer molecules. For this reason I consider that the results from experiments with succinic acid in this study are not valid to derive conclusions on the average behavior of marine biogenic organics.*

b) *Some of the succinic acid concentrations applied in this study are much higher than what would be expected to find in real seawater samples. The authors indicate that 94 uMol/L is a representative concentration of DOC in Arctic ocean seawater but 4 orders of magnitude above the average concentration of succinic acid in seawater. Nevertheless, concentrations up to 2450 umol/L are applied in their experiments. For comparison purposes it is fine to use a wide range of concentrations; however the results obtained for these high concentrations are not valid to derive conclusions for real conditions. It is misleading to state in the abstract that succinic acid produces a 43% reduction in the particle production, when this is what happens for the 2450 umol/L case. Statements in abstract and conclusions should be limited to findings that can be extrapolated to real conditions.*

1. Reply:
a) We thank the reviewer for this comment. We do not want to claim that succinic acid represents the average behavior of marine biogenic organics. We are aware that the use of bioexudates would have been a better choice. The selection of the surfactant used in the experiments was limited by the availability of suitable compounds. Our ambition was not to fully simulate the role of organic matter in PMA production, but more to show the general trend how they influence the PMA production at low temperatures. Do they enhance the increase of PMA emissions with decreasing sea water temperature or not? For this purpose we believe that using succinic acid as proxy was a sufficient choice. We missed to point out that succinic acid although it is synthesized and metabolized in sea water is only representing a small group of surfactants. We changed the manuscript in the way that we mention the difference between properties of succinic acid and biogenic polymers and point out that succinic acid is not appropriate for representing average marine biogenic organics.

b) We changed the abstract and the summary section and included mainly results of the experiment when water with a succinic acid concentration of 94µmol l^{-1} was used. However, the cooling/warming experiment with water containing succinic acid was only conducted for a concentration of 2446 µmol l^{-1}. This was so, as our aim was amongst others, to test the impact on the water temperature dependent trend of particle number concentration by exaggerating to clarify the impact of a change in water temperature compared to changes in other parameters. Therefore, the highest succinic acid concentration was used in the cooling/warming experiment (Fig. 2 and Fig. 3).

1. Revision:
a) Section 2.3 was changed from:
"The surfactant chosen for the experiments was succinic acid, since it is a water-soluble organic compound and ubiquitous in different compartments of the environment (Mahiuddin et al., 2008; Hyvärinen et al., 2006). Succinic acid belongs to a group of low molecular weight dicarboxylic acids (DCA), which are amongst others synthesized and metabolized in different biochemical processes in sea water (Steinberg and Bada, 1984; Kester and Foster, 1963). Succinic acid is, with concentrations of less than 1 µg l^{-1} one of the lower concentrated DCA in sea water (Tedetti et al., 2006), but can still be considered to represent other DCA, such as oxalic acid, malonic acid and adipic acid." (lines 21-26, p 19043; lines 1-3, p 19044) to:

"The surfactant chosen for the experiments was succinic acid, since it is a water-soluble organic compound and ubiquitous in different compartments of the environment (Mahiuddin et al., 2008; Hyvärinen et al., 2006). Succinic acid belongs to a group of low molecular weight dicarboxylic acids (DCA), which are amongst others synthesized and metabolized in different biochemical processes in sea water (Steinberg and Bada, 1984; Kester and Foster, 1963). Succinic acid is, with concentrations of less than 1 µg l^{-1} one of the lower concentrated DCA in sea water (Tedetti et al., 2006), but can still be considered to represent other DCA, such as oxalic acid, malonic acid and adipic acid. **Even that succinic acid is present in the ocean, it cannot be considered to represent the behavior of an average marine biogenic organic. The exudates of marine organisms like phytoplankton and bacteria, are characterized by polymeric structures which eventually may assemble and form microgels (Chin et al., 1998; Passow, 2002). The polymeric structures and microgels are characterized by high molecular weights and therefore next to their difference in chemical properties have a different impact on the physical properties of water compared to succinic acid. Although succinic acid is not representing the impact of average biogenic organics on water properties and consequently on aerosol properties, it is justifiable to use it as a proxy for the experiments carried out in this study.**"

b) To specify the particle number concentration change with a change in succinic acid concentration for different sizes, 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 1b) 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}.**"

The 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 %.**"

2. Comment:
The authors should make clear that the fact that the Arctic seawater distributions in Fig. 10 were shifted towards larger sizes with respect to the pure NaCl measurements could also be due to the presence of inorganic compounds other than NaCl. Thus, although the succinic acid and Arctic seawater results were both shifted with respect to the pure NaCl measurements, it is not demonstrated that this is due to marine organics in the Arctic seawater nor that succinic acid can be used as a proxy for these organics.

2. Reply:
We agree with the referee and rephrased a sentence in the Discussion and Conclusion Section.
The sentence:

"It should however be noted that the effect of succinic acid qualitatively agree better with what we found for the complex mixture of organics in the real ocean water, while the effect described by Sellegri et al. (2006) for SDS was the opposite." (lines 11-14, p19060) was changed to:

"**Although, it may seem that the effect of succinic acid qualitatively agrees better with what we found for the complex mixture of organics in the real ocean water (compared to the use of SDS by Sellegri et al., 2006), it has to be considered that the size distribution based on Arctic Ocean water is also likely influenced by the presence of other inorganic substances than NaCl.**"

In the Summary one bullet is slightly modified. The sentence:
"High concentrations of succinic acid and other unknown organics and salts present in Arctic Ocean water alter the size distribution resulting of pure NaCl water. A shift to larger sizes is observed from pure NaCl water to Arctic Ocean water." (lines 20-23, p19066) was changed to:
"High concentrations of succinic acid and other unknown organic **components** and **inorganic** salts present in Arctic Ocean water alter the size distribution resulting of pure NaCl water. A shift to larger sizes is observed from pure NaCl water to Arctic Ocean water."

3. Comment:
Section 5.2- Previous studies on the influence of organics on particle production by Sellegri et al., Fuentes et al. and Tyree et al. showed a shift of size distributions toward smaller sizes or an increase in particle production with increasing organic concentration regardless of the experimental set up used. The authors claim that the different behavior observed in their measurements with succinic acid is likely due to the different experimental set-up and organic compound used. The authors should note that previous works were conducted at room temperature, while measurements in this paper are performed at very low temperature. Surfactants behavior is indeed strongly affected by temperature and this could be key to explain why a different trend was observed in this study with respect to previous literature.

3. Reply:
Thank you for pointing out the water temperature difference between the experiments. Some sentences concerning the water temperature dependence on surfactant behavior will be added.

3. Revision:
After the sentence:
"Fuentes et al. (2010b) used the exudates of algae present in high biological active regions on the west coast of Africa, which is presumably different to the late winter Arctic Ocean water sample used in our experiments." (lines 17-20, p19061) some sentences concerning the water temperature and the impact on surfactants are added:
"**Although different kind of surfactants used in the present study, the study by Sellegri et al. (2006), Tyree et al. (2007) and Fuentes et al. (2010b) makes it difficult to compare the results, it is important to mention that water temperature may have impacted as well on the surfactant's properties. While the presented succinic acid experiments were conducted**

with very low water temperatures around 0 °C, the other studies were conducted with waters having temperatures above 20 °C. It was shown that the critical micelle concentration in the water is a function of water temperature (Mehta et al., 2005; Chen et al., 1998). In addition it was demonstrated that already small changes of temperature can change the size, density, dielectrical proberties, and chemical reactivities of gels (Tanaka et al. (1992) in Verdugo et al., 2004). "

Additionally the sentence:
"As shown previously, a comparison between laboratory experiments is difficult due to different aerosol generation mechanisms, different waters and different organic substances used."(lines 8-10, p19065) is changed to:
"As shown previously, a comparison between laboratory experiments is difficult due to different aerosol generation mechanisms, different waters**, partly different water temperatures,** and different organic substances used."

4. Comment:
As a last comment it is important that the authors acknowledge not only a need for reaching a consensus on the type of experimental set-up to be used in different studies but also on using representative proxy biogenic organic mixtures covering a wide range of molar masses rather than individual low molar mass compounds.

4. Reply:
We agree with the referee and augmented the manuscript accordingly to the comment.

4. Revision:
The sentence:
"More studies are needed for examining the influence of water temperature, salinity, soluble and insoluble organic substances present in different marine environments on particle number production by using only one experimental setup." (lines10-13, p19065) is augmented to:

"More studies are needed for examining the influence of water temperature, salinity, soluble and insoluble organic substances present in different marine environments on particle number production by using only one experimental setup. **Particular attention should be paid to the choice of the surfactant. A representative proxy for biogenic organic mixtures covering a wide range of molar masses and so a large variety in chemical and physical properties, should be chosen.** "

Minor comments

1. + 2. Comment:
Abstract Line 7: replace "smaller" by "small".
Line 7: this sentence needs rephrasing; I would suggest the following "from 0 C where they represent 85-90% of the total aerosol number to 10 C, where they represent 60-70% of the total number".

1. + 2.Reply:
The replacement and rephrasing was done. (Abstract line 7)

1. + 2. Revision:
"**Small** particles with a dry diameter between 0.01 µm and 0.25 µm dominate the aerosol number density, but their relative dominance decreases with increasing water temperature **from 0 ◦C where they represent 85–90 % of the total aerosol number to 10 ◦C, where they represent 60–70 % of the total aerosol number**."

3. Comment:
Statements in lines 7-10 and 12-14 in the abstract seem contradictory. In lines 7-10 it is said that there is a reduction in the particle number from 0 to 10 C, while in lines 12-14 it is stated that there is no change in the shape of the particle size distribution in the temperature range between 0 and 16 C.

3. Reply:
Thank you for pointing that misleading formulation out.

3. Revision:
To eliminate the contradictive statements, the sentence:
"A change of salinity between 15 g kg^{-1} and 35 g kg^{-1} showed no influence on the relative shape of a particle number size distribution, nor did a change in water temperature between 0 ◦C and 16 ◦C."(line 12-14, p19040) was rephrased to:

"**A change of salinity between 15 g kg^{-1} and 35 g kg^{-1} did not influence the shape of a particle number size distribution. Although the magnitude of the size distribution for a water temperature change between 0 ◦C and 16 ◦C changed, the shape did not.**"

4. Comment:
I find it difficult to follow the last sentence in the abstract. Is it meant here that the distributions with succinic acid and Arctic sea water were shifted towards larger sizes with respect to the NaCl experiments? 19043 Lines 20-29. Please modify this section taking into account the discussion presented in the major comments.

4. Reply:
The last sentence in the abstract is rephrased. Based on Major Comment 1b), the water having a succinic acid concentration of 2446 µmol l^{-1} will not be considered.

4. Revision:
The sentence:
"Different organic constituents and perhaps inorganic substances resulted in a particle number shift towards larger particle sizes, when comparing a size distribution resulting from pure NaCl water to size distributions resulting from Arctic Ocean water and resulting from NaCl water with a succinic acid concentration of 2446 µmol l^{-1}" (lines17-21, p19040) is rephrased to:
"**A shift to larger sizes in the particle number size distribution is observed from pure NaCl water to Arctic Ocean water. This is likely a consequence of organics and different inorganic salts present in Arctic Ocean water in addition to the NaCl.**"

5. Comment:
19045 line 9: replace "to minimize any influence of organic matter" by "to minimise any organic matter contamination"

5. Reply:
The sentence in lines 9-11, p 19045 was rephrased.

5. Revision:
"To minimi**s**e any **organic matter contamination**, salinities of 18 g kg^{-1} and 35 g kg^{-1} were achieved by adding NaCl (sodium chloride, puriss.p.a, Sigma-Aldrich) to fresh MQ water in the buffer tank."

6. Comment:
19060 lines 11-14. The fact that the Arctic seawater distributions were shifted towards larger sizes with respect to the pure NaCl measurements could also be due to the presence of other inorganic compounds. It is misleading that the authors state that similar behavior was obtained for the succinic acid and Arctic waters measurements, while the effect of other inorganic substances could have affected the results obtained for the Arctic waters.

6. Reply:
We agree with the referee and rephrased the sentence.

6. Revision:
The sentence:
"It should however be noted that the effect of succinic acid qualitatively agree better with what we found for the complex mixture of organics in the real ocean water, while the effect described by Sellegri et al. (2006) for SDS was the opposite." (lines 11-14, p19060) was changed to:

"**Although, it may seem that the effect of succinic acid qualitatively agrees better with what we found for the complex mixture of organics in the real ocean water (compared to the use of SDS by Sellegri et al., 2006), it has to be considered that the size distribution based on Arctic Ocean water is likely influenced by the presence of other inorganic substances than NaCl.**"

References:

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