

# Author's response on the revised manuscript: Dimethylsulfide dynamics in first-year sea ice melt ponds in the Canadian Arctic Archipelago

Margaux Gourdal<sup>1</sup>, Martine Lizotte<sup>1</sup>, Guillaume Massé<sup>1</sup>, Michel Gosselin<sup>2</sup>, Michel Poulin<sup>4</sup>, Michael  
5 Scarratt<sup>3</sup>, Joannie Charette<sup>2</sup>, Maurice Levasseur<sup>1</sup>

<sup>1</sup>Département de biologie, Québec-Océan and Unité Mixte Internationale 3376 TAKUVIK, CNRS-Université Laval, 1045  
avenue de la Médecine, Québec, Québec G1V 0A6, Canada

<sup>2</sup>Institut des Sciences de la Mer de Rimouski (ISMER), Université du Québec à Rimouski, 310 allée des Ursulines,  
10 Rimouski, Québec G5L 3A1, Canada

<sup>3</sup>Maurice Lamontagne Institute, Fisheries and Oceans Canada, P.O. Box 1000, Mont-Joli, Québec G5H 3Z4, Canada

<sup>4</sup>Research and Collections, Canadian Museum of Nature, P.O. Box 3443, Station D, Ottawa, Ontario K1P 6P4, Canada

*Correspondence to:* Margaux Gourdal (margaux.gourdal@takuvik.ulaval.ca)

## Copyright statement

15 © Author(s) 2017. CC-BY 3.0 License.

The author's response is structured as follows: (1) comments from referee #1, with the associated author's response and author's changes in manuscript (pp. 2 – 8); (2) comments from referee #2, with the associated author's response and author's changes in manuscript (pp. 8 – 31); (3) a marked-up manuscript version is then presented. (p. 32-80)

#### **General comments of anonymous referee #1 :**

5 Referee #1: This is a generally well written and interesting manuscript describing novel measurements of Dimethylsulfide (DMS, DMSP<sub>d</sub> and DMSP<sub>p</sub>) concentrations and dynamics (derived from labelled DMSP and DMSO isotopic marker incubations) in Arctic sea ice melt ponds. A shortcoming of the paper is that it is based on a rather limited dataset with consequent problems for statistical analyses. Only two (brackish) melt ponds were sampled for incubations in this study, and statistics are based on an N=2 (with additional duplicate - but apparently dependent - samples taken from each incubation).  
10 While a t-test can be employed for a dataset with an N=2(4), the dataset appears extremely small to make any statistical relevant conclusions. This reviewer therefore suggests to clarify (provide df or define N values) or alternatively delete these statistical analyses and rephrase some of the statements in relation to DMSP transformation into DMS. This said, other methods applied in this study appear to be solid (noting that this reviewer is not an expert in GC/GC-MS DMS(P) analyses) and raise some important new research questions for future research on DMS dynamics in sea ice melt ponds. In summary  
15 this reviewer suggests publication of the manuscript after amending the statistical analyses (t-test) and some other (minor) shortcomings including a re-consideration of the estimate of the overall DMS reservoir in Arctic melt-ponds, and a more detailed discussion on the sea ice surface permeability.

#### **Author's response to general comments of anonymous referee #1 :**

20 We thank the reviewer for his/her positive general evaluation of the paper and helpful comments. The following actions were taken:

-We acknowledge that our restricted dataset limited the power of the chosen statistics analyses. We thus removed the results of the Student's t-test (P8, L25) and used a non-parametric Mann-Whitney U test (a replacement for independent groups t-test) that allowed us to compare the two independent groups of samples from the Ice1-MPI and Ice4-MP1 incubation experiments. The Mann-Whitney U test revealed no significant differences between the distributions of the  
25 reduced-sulfur compounds (i.e. DMS, DMSP<sub>d</sub> and DMSP<sub>p</sub>) from the Ice1-MP1 and Ice4-MP1 incubation results (n=45, df=16,  $\alpha=0.05$ ). The conclusions from this first step warranted the combination of the Ice1-MP1 and Ice4-MP1 datasets resulting in both greater sample size and statistical power for further analyses.

-Based on the results of the Mann-Whitney U test, the second step involved using a series of Wilcoxon Signed-rank tests on the combined datasets in order to 1) assess the presence of statistical differences between the Controls and each  
30 Treatment L-DMSP/O and D-DMSP/O; 2) assess the potential effect of light on the concentrations and change rates of the reduced sulfur compounds under study (DMS, DMSP<sub>d</sub> and DMSP<sub>p</sub>) by comparing paired dependent samples (repeated

measures) from L-DMSP/O and D-DMSP/O. As recommended, df values are now provided for each statistical test performed.

- We deleted the section of the discussion where we tentatively estimated the overall size of the DMS reservoir in Arctic melt-ponds. We agree that a greater spatial coverage of MPs is needed to come up with a more robust estimate.

5 - In the initial submission, brine volumes were calculated from the T–S measurements. In the revised version, full depth temperature, salinity and brine volume profiles in sea ice are presented in a new figure (new Figure 3; the “figure 3” previously presented became “figure 4” in the revised version of the manuscript) for stations Ice1, Ice3 and Ice4 in support of a more detailed discussion on the sea-ice surface permeability.

10 Caption of figure 3: In situ temperature (●) and bulk ice salinity (○) profiles of the sea ice surrounding the melt ponds sampled at stations Ice1 (a), Ice3 (b) and Ice4 (c). Temperature and salinity values of each 10 cm sea ice section were used to calculate brine volumes (■) throughout the full depth of sea ice, an indicator of sea ice permeability.

#### **Author’s response to specific comments of anonymous referee #1:**

**R1: P2, L1:** delete “natural” in first sentence of abstract, this word is not needed.

15 **Response:** Done.

**R1: P2, L12:** This calculation of the DMS reservoir in Arctic melt-ponds is based on 2 single measurements of 2 very specific (=brackish) melt ponds in a very defined study area (e.g. the Canadian Archipelago). This reviewer considers up-scaling the results from this study to the entire Arctic as highly problematic. It is suggested to delete this estimate from the manuscript (see also page 16) or at least to delete this broad-brush estimate from the Abstract.

20 **Response:** We deleted the calculation of the size of the DMS reservoir in Arctic FYI melt-ponds from the manuscript.

**R1: P4, L17:** No need to start a new paragraph.

**Response:** This paragraph was merged with the previous paragraph.

25

**R1: P4, L32:** be more specific: “of melted ice samples” rather than “melt water samples”

**Response:** The sentence now states “of melted ice [...]”.

**R1: P5, L1:** The T and S data from the 10 cm surface ice allow the accurate calculation

of the brine volume fraction according to established formulas, see e.g., Eicken, H., H. R. Krouse, D. Kadko, and D. K. Perovich, Tracer studies of pathways and rates of meltwater transport through Arctic summer sea ice, *J. Geophys. Res.*, 107(C10), 8046, doi:10.1029/2000JC000583, 2002; an references therein. Applying these formulas (e.g. those for high T and low S sea ice values, e.g. Manninen and Leppaeranta 1988, cited in above reference), and  
5 using the values reported in the manuscript of  $T = -0.2\text{C}$  and  $S = 0$  psu actually indicates “im”-permeable ice, while a  $T = -0.2\text{C}$  and  $S = 0.8$  psu indicates a brine volume of about 20% (= highly permeable ice). This reviewer suggest that brine volumes are calculated for the T – S measurements and that a more detailed discussion on ice permeability/sea water percolation is given. Please also note that a) “the rule of 5s” is primarily based on a brine volume fraction of 5% (which can be achieved by different T-S combinations, including  $T = -5\text{C}$  and  $S = 5$ , b) that this percolation threshold is only valid  
10 during thermodynamic equilibrium, and c) also only applies for columnar ice (likely the case in these samples), but this surface ice might have also undergone some melting/metamorphosis). In summary this reviewers suggest a more detailed discussion of the sea ice permeability. The current conclusions are fine, but just stating “according to the rule of 5s” is insufficient.

**Response:** A more detailed discussion on ice permeability/sea water percolation is now included in the manuscript.  
15 Full ice depth T and S profiles are now presented in figure 4 for stations Ice1, Ice3 and Ice4, and brine volumes were calculated from the T–S measurements.

The method section was changed accordingly (P4, L28) and now states that :

“In order to estimate the possibility of a connexion between the melt ponds sampled and the underlying sea ice (i.e. through ice permeability or water percolation), sea-ice salinity and temperature were measured. For each station where sea  
20 ice was sampled, an in situ sea-ice temperature profile was measured directly, at 0.1 m intervals, using a high-precision thermometer (Testo 720). Corresponding sea-ice salinity profiles were also determined at 0.1 m intervals. Each 0.1 m section was cut with a handsaw, stored in a plastic container, and allowed to melt at room temperature. Bulk salinity of the melted ice section was determined using a conductivity probe (Cond 330i, WTW). Brine volume profiles were calculated using the recorded sea-ice bulk salinity and in situ temperature (Cox and Weeks 1983, Petrich and Eicken 2010)”.

25 Consequently, the result section was also modified (P9, L5). The text now reads:

“Averaged values for bulk sea-ice salinity over the full thickness of the ice were 1.73, 2.83 and 3.75 at stations Ice1, Ice3 and Ice4, respectively. Maximum bulk salinity never exceeded 5.00 (Ice4, 1.2-1.3 m section). In situ temperatures, averaged over the full thickness of the ice, were  $-0.54\text{ }^{\circ}\text{C}$ ,  $-0.52\text{ }^{\circ}\text{C}$  and  $-0.98\text{ }^{\circ}\text{C}$  at stations Ice1, Ice3 and Ice4, respectively, and reached a minimum value of  $-1.39\text{ }^{\circ}\text{C}$  (Ice4, 0.8-0.9 m section). Brine volume fraction constantly exceeded 10% in the ice  
30 profiles, except in the upper 0.1 m section of the Ice3 station.”

**R1: P5, L 11:** “replicates” How many?

**Response:** This was changed to “duplicates”.

**R1: P5, L25:** This reviewer suggest to add a sentence and a definition of “HNA” here, e.g.  
5 what nucleic acid stain was used in this fly cytometry protocol?

**Response:** The following sentence was added to the text: “Heterotrophic bacteria samples were stained with SYBR  
Green I and measured at 525 nm to quantify bacteria with Low Nucleic Acid (LNA; potentially less active) and High  
Nucleic Acid (HNA; potentially more active) content (Gasol and del Giorgio 2000, Lebaron et al. 2001). Analyses were  
performed on an Epics Altra flow cytometer (Beckman Coulter), fitted with a 488 nm laser (15 mW output; blue), using  
10 Expo32 v1.2b software (Beckman Coulter).”

**R1: P6, L23:** It is unusual to refer to PAR as “700-400”, normally one would write “400-700”.  
This also applies to the UVA and UVB wavelengths given in the text.

**Response:** All the wavelengths presented in the text are now written in the suggested format.

15

**R1: P 8, L25:** As discussed above, this reviewer suggests to revisit the t-test statistics applied: It appears that N  
equals 2, which makes application of the t-test problematic. At least more explanation is needed.

**Response:** As recommended, results from the Student's t-test are no longer presented. We nevertheless wanted to  
base our analysis on statistical tests. To do so, we explored the possibility of pooling our incubations data in order to increase  
20 ‘n’. A non-parametric Mann-Whitney U test was first used to determine whether the distributions of reduced-sulfur  
compounds (i.e. DMS, DMSP<sub>p</sub> and DMSP<sub>d</sub>) in the Ice1-MP1 and Ice4-MP1 incubations experiments were statistically  
different from one another. The difference in reduced-sulfur compound concentrations between the two incubation  
experiments was not found to be statistically significant (n=45, df=16  $\alpha=0.05$ ).

As explained previously in the general comments section, this allows us to combine the results of Ice1-MP1 and Ice4-MP1  
25 when testing for differences in responses between Treatments. This doubling of sample size (n) for each test (combining  
Ice1-MP1 and Ice4-MP1) led to an increase of the statistical power of the analysis conducted hereafter. A Wilcoxon Signed-  
rank test was used to assess potential statistical differences between the Controls and each Treatment L-DMSP/O and D-  
DMSP/O. Results reveal significant differences ( $p\leq 0.05$ ) between the Controls and each Treatment of the incubation  
experiments (n=30, df=8,  $\alpha=0.05$ ). Further detail on the other statistical tests conducted is provided in the response to the  
30 “(P11, L10 -15)” comment.

**R1: P 9, L3:** This reviewer suggest to use the SI unit “m” rather than “cm” as unit for length measurements throughout the manuscript/figures.

**Response:** “cm” was replaced with SI unit “m” throughout the manuscript and figures.

5 **R1: P9, L7:** As per above more details is required than just stating the “rule of fives”.

**Response:** More details are provided for this section in the response to the “P5, L1:” comment.

**R1: P9, L 16:** use singular, e.g. “detail”

**Response:** The singular is now used in the text.

10

**R1: P11, L10 -15:** If “significantly” is used test-statistics should be given, also provide df value and/or N. Given the low N, these statistical results are of little relevance.

15 **Response:** Each statement of the paragraph (between quotation marks) is now followed by a description of the statistical test used. As stated previously, results of Ice1-MP1 and Ice4-MP1 were combined (as justified by the the results of the Mann-Whitney U test), resulting in an increase of the statistical power of the analysis conducted.

“During both Ice1-MP1 and Ice4-MP1 incubation experiments, the light Treatment had no effect on the net changes in DMSP<sub>d</sub> concentrations between the L-DMSP/O and D-DMSP/O Treatments [...]”.

→This was assessed using a Wilcoxon Signed-rank test (n=8, df=3,  $\alpha=0.05$ ) comparing pairwise DMSP<sub>d</sub> concentrations at T<sub>6</sub>, T<sub>12</sub>, T<sub>18</sub>, and T<sub>24</sub> for both incubation experiments Ice1-MP1 and Ice4-MP1,  $p\geq 0.05$  .

20 “[...] But significantly impacted the rates of net accumulation of DMS”

→ This was assessed using a Wilcoxon Signed-rank test (n=12, df=5,  $\alpha=0.05$ ) with a significance level of  $p\leq 0.05$  comparing pairwise the DMS accumulation rates in L-DMSP/O versus D-DMSP/O at T<sub>0</sub>-T<sub>6</sub>, T<sub>6</sub>-T<sub>12</sub>, T<sub>12</sub>-T<sub>18</sub>, T<sub>18</sub>-T<sub>24</sub>, T<sub>6</sub>-T<sub>24</sub> and daily rates (T<sub>0</sub>-T<sub>24</sub>) for both incubation experiments Ice1-MP1 and Ice4-MP1.

25 “The accumulation of DMS over 24h in the L-DMSP/O Treatments were consistently and significantly lower than in the corresponding D-DMSP/O Treatments ( $p\leq 0.05$ ) (Fig. 3b, d).”

→ This was assessed using a Wilcoxon Signed-rank test (n=8, df=3,  $\alpha=0.05$ ) comparing pairwise DMS concentrations in L-DMSP/O versus D-DMSP/O at T<sub>6</sub>-T<sub>12</sub>-T<sub>18</sub> and T<sub>24</sub> in both incubation experiments Ice1-MP1 and Ice4-MP1.

**R1: P 12, L 17:** Sea “spray” rather than “spay”

**Response:** Typo corrected.

**AR1: P12, L28:** Here “gravity drainage” and “brine flushing” are used to describe the same process, while classically “brine drainage” refers to the release of cold salt brines in surface-cooled sea ice, while “brine flushing” refers to the flushing out of salt through meltwater, e.g. they are technical terms used for different physical processes.

**Response:** The technical terms are now correctly used in the text. The discussion on the salt movements through sea ice has also been amended. The corrected section is described below:

(P12, L27) “It is also unlikely that sea-ice brine intrusion contributed to the salinization of the melt ponds since the ponded FYI sampled in this study appears to be almost fresh (using the terminology proposed in Vancoppenolle et al., 2007) (Figure 3). Consolidated cold FYI generally exhibits a characteristic C-shaped salinity profile (Nakawo and Sinha, 1981) after loosing approximately two thirds of the initial seawater salt content through gravity drainage in winter (Kovacs, 1996). Then, according to the mushy-layer theoretical representation of sea ice, most of the salted brines are usually lost through full depth brine convection well before melt ponds start to form (Jardon et al., 2013). Finally, residual salts are lost during brine flushing events, typical of the summer season (Weeks and Ackley, 1986, Eicken et al., 2002; Vancoppenolle et al., 2007). The low salinity values and the flattened salinity profile observed in the sampled sea ice suggest that the ice had already been subjected to brine flushing. We thus exclude sea-ice brine enrichment of melt ponds as a significant salinization mechanism”.

(P13, L3): “This leaves seawater intrusion through highly porous sea ice as the most likely process responsible for bringing salts, microorganisms, and DMS in melt ponds. Above a brine volume threshold of 5%, sea ice becomes permeable to fluid transport through its interconnected brine network (Golden et al., 1998). Melt ponds form and persist despite the high porosity of FYI due to the infiltration and subsequent freezing of a freshwater layer into the pore structure of sea ice that prevents percolation drainage of pond meltwater (Polashenski et al., 2017). Here, the brine volume fraction calculated for each 0.1 m section always exceeds 10%, suggesting that sea ice was highly permeable throughout the full ice depth (except for the upper 0.1 m of Ice3). As brines flushes out of the ice, seawater fills the channel network (Widell et al., 2006). Some degree of connectivity is thus expected to take place between superficial melt ponds and seawater. Specifying whether the intruding seawater originates from lateral or direct upward flow is difficult since these processes are not yet well understood (Vancoppenolle et al., 2007). Sea-ice freeboard was either low or negative near the melt ponds sampled (Table 1), suggesting that seawater intrusion through highly porous low-freeboard sea ice was possible in the observed sea ice. The somewhat higher freeboard measured at station Ice3 *may indicate refreezing metamorphosis of snow*. Sea-ice recrystallization could explain the impermeability of the upper 0.1 m of sea ice at station Ice3. The low-freeboard configuration at stations Ice1 and Ice4 is the general fate of melting sea ice, and inherent to the loss of sea-ice thickness. Our hypothesis of seawater intrusion

through highly porous low-freeboard sea ice is also supported by the presence of both pelagic and ice-associated algae in the microbial assemblages of the melt ponds, along with the similarity observed between algal species composition in the waters of the melt ponds and those beneath the ice (Charette et al., personal comm.). The seeding of these seawater microorganisms into melt ponds may also affect the cycling of DMS as discussed in sect. 4.2.”

5

**P 13, L 10:** No data are shown that demonstrate: “full depth desalinization” -> please clarify

**Response:** This statement was removed from the manuscript. Calculations provided by Jardon et al. (2013) deal with the permeability threshold of sea ice with salinity greater than 5 psu. With a bulk sea ice salinity of 2.79 (averaged for the three stations), with a maximum value of 5.00 at station ice4 (1.2-1.3 m section), we fall outside of this range.

10

**R1: P 13, L 20:** Avoid the use of “significant” if no statistical test was conducted /or provide statistical results.

**Response:** This was changed to “A daily net DMS production [...]”.

Fig and Tables:

15

**R1: Fig 3:** Unusual numbering of panels: “c” should be “b” and “b” should be “c”?

**Response:** The numbering was changed as suggested.

**R1: Tab 7:** “control” or “Control” -> consistency in spelling needed

20

**Response:** The consistency of “control” spelling was checked and applied throughout the text.

### **General comments of referee #2 :**

Referee G. Carnat: The study of Gourdal et al. discuss the dynamics of the climate-active gas dimethylsulfide (DMS) in surface melt-ponds developing over Arctic first-year sea ice. The authors present an original data set of DMS(P) concentrations measured in nine melt-ponds combined with ancillary physical and biological parameters. Based on these data, the authors discuss several physical processes to explain the presence of DMS and microbial organisms in the melt-ponds. Then, the authors use incubations with stable isotope-labelled DMSP and DMSO to investigate de novo biological production of DMS in the melt-ponds via different pathways. As mentioned by the authors, this study represents the first effort to characterize the cycling of DMS in Arctic melt-ponds, an interesting medium at the interface between sea ice and the atmosphere which importance is expected to increase in the future. Overall, the paper is well organized and well written.

30



I would say that the methods regarding the DMS,P concentrations, incubations with isotopes, and ancillary biological parameters are adequate and well described. The DMS,P data, especially the results from the incubations experiments, are well presented and discussed in a very convincing way. That being said, I think that the physical component of the melt-pond/sea ice system is on the other hand poorly constrained in the study. There are numerous errors and approximation in each section of the manuscript regarding for instance sea ice permeability. I provided multiple suggestions and corrections in the specific comments detailed below and I strongly encourage the authors to follow these suggestions. This is my main criticism on the paper and I think this part should be improved before publication. I identified two other minor shortcomings. First, I think that the DMS cycling in melt-ponds could be better put in the general context of the DMS sea ice cycling, especially in the introduction. Second, I think that not enough precautions are taken when the regional estimates of the contribution of melt-ponds in the DMS cycle is assessed in the manuscript given the relative small number of samples considered. Also, this contribution should be compared to oceanic and sea ice contributions. Listed below are additional small and specific comments and recommendations. In summary, I suggest publication of the manuscript once the three (minor) issues identified above have been tackled and specific comments addressed.

#### **Author's response to general comments of referee #2 :**

We thank the reviewer for his positive general evaluation of the paper and the insightful comments. The following general actions were taken:

- The discussion on the physical component of the melt-pond/sea ice system has been extensively amended following the suggestion of the reviewer. The discussion on sea ice permeability is now supported by brine volume calculations. We carefully considered the suggestion to use the Rayleigh number (Ra) in our analysis of ice permeability, however we finally decided to exclude this parameter. Details are provided in the supplementary material joined to our response to the reviewer. Briefly, we expect large uncertainty of Ra number calculation (VanCoppennolle et al., 2013) because brine loss during sampling of highly permeable sea ice, conditions encountered during our sampling period, has been shown to lead to an underestimation of bulk salinity (Notz et al., 2005) .

- Several changes were made to the introduction section, as suggested by the reviewer. These changes include 1) a more detailed introduction of the DMS cycling (including more information on DMSO), 2) a clarified comparison with Antarctic melt ponds, and 3) a better description of the gap addressed by our study.

-We deleted the estimate of the size of DMS reservoir in Arctic FYI melt ponds from the manuscript. We agree that small datasets, such as the one presented in our study, carry inherent limitations that make extrapolation calculations difficult.

#### **Author's response to specific comments of referee #2:**

**P2, L2** (and throughout the manuscript): “first-year” instead of “first year”.

**Response:** This was corrected throughout the manuscript.

5 **P2, L3** (and throughout the manuscript): sea ice instead of sea-ice. Please be consistent throughout the manuscript.

**Response:** This was corrected throughout the manuscript.

**P2, L6:** “In the Eastern Canadian Arctic”, I would use “Canadian Arctic Archipelago” to be consistent with the title.

10 **Response:** We changed the sentence to fit the title formulation.

**P2, L7:** Please check throughout the manuscript that “ca.” is the proper scientific notation. Also, you could provide a range and standard deviation here between brackets.

15 **Response:** We did not find any Biogeosciences Discussions (BGD) guideline regarding the “ca.” notation, but we checked other BGD papers. All of them used the tilde symbol “~” and not “ca.”. We now use the tilde symbol the manuscript.

The text now reads as “[...] and increased linearly with salinity ( $r_s = 0.84$ ,  $p \leq 0.05$ ) from 2.6 up to 6.1 nmol l<sup>-1</sup> (avg.  $3.7 \pm 1.6$  nmol l<sup>-1</sup>) in brackish melt ponds.”

20 **P2, L9:** “Experiments conducted with” rather than “Results from experiments”. This is a little bit redundant with the next sentence.

**Response:** The sentence starts now with “Experiments conducted with [...]”.

**P2, L10:** Bracket missing here.

25 **Response:** A bracket was added.

**P2, L11-15:** As explained in my general comments on the paper and on the conclusion, I think you should be a little bit more careful with this sentence since it is based on a very limited number of samples taken in a very limited area of the Arctic. While I believe it fits well in the conclusion where you have room to develop on limitations and future work to be conducted, you might want to remove it from the abstract. It is definitely not the key message of your paper. Should you  
5 keep it, I would at least put your estimate in perspective compared to other potential sources (open water, leads, sea ice itself, ...).As it is, it is not clear for the reader if melt-ponds are a small or significant reservoir of DMS.

**Response:** We agree that small datasets such as presented in our study carry inherent limitations that make extrapolation calculations difficult. We therefore decided to delete the estimate of the size of DMS reservoir in Arctic FYI melt ponds from the manuscript.

10

**P3, L12:** “DMS-derived sulfate aerosols”.

**Response:** The word “sulfate” was added to the text.

**P3, L14:** Please indicate the two different backscattering effects of DMS-derived sulfate aerosols (direct and  
15 indirect through CCN).

**Response:** Please see the re-written paragraph in the P3, L11-15 response below.

**P3, L11-15:** Please introduce here quickly the controversy about the CLAW hypothesis (cfr. e.g. Quinn and Bates, 2011, Green and Hatton, 2014) and the influence of DMS on a global scale. Then you can make the connection to the next  
20 sentence and talk about the influence of DMS on a more regional scale.

**Response:** The paragraph starting (P3, L11) was modified according to the suggestions P3, L12 ; P3, L14 ; P3, L11-15 ; P3, L16 ; P3, L19 ; P3, L20:

The paragraph now reads: “Dimethylsulfide (DMS) is the main natural source of reduced sulfur for the atmosphere (Bates et al., 1992). Between 17.6 to 34.4 Tg of sulfur are released annually from the ocean to the atmosphere (Lana et al., 2011),  
25 accounting for 50-60% of the natural reduced sulfur emitted (Stefels et al., 2007). DMS is also a climate-relevant gas potentially involved in a feedback loop known as the “CLAW” hypothesis (Charlson et al., 1987) linking biology and climate through the production of DMS-derived sulfate aerosols. According to CLAW, DMS emissions may affect the global radiation budget directly through the scattering of incoming solar radiation, and indirectly via the production of cloud condensation nuclei (CCN) leading to the genesis of longer-lived clouds with higher albedo (Twomey, 1974; Albrecht,  
30 1989). Inspiring three decades of research and hundreds of publications, the feedback mechanism proposed by Charlson et

al. (1984) remains yet to be demonstrated in its entirety (e.g. Ayers and Cainey, 2008 ). Although modelling results show that DMS emissions may have a negative radiative effect (e.g. Bopp et al., 2004; Gunson et al., 2006; Thomas et al., 2010), CCN may exhibit a low sensitivity to changes in DMS on a global scale (Woodhouse et al., 2010). Recent studies questioning the relative importance of DMS in new particle formation have emerged, suggesting that the global CLAW  
5 feedback may be weak (e.g. Quinn and Bates, 2011; Green and Hatton, 2014). On a regional scale however, the response of CCN production to change in DMS may vary by a factor of 20 (Woodhouse et al., 2010). The impact of DMS emissions on cloud properties (through the production of CCN) could be particularly important in remote pristine marine areas such as the polar regions (Carslaw et al., 2013). In the Southern Ocean, DMS may have contributed up to 33% of the increase in CCN observed south of 65°S as a response of increased wind speed since the early 1980s (Korhonen et al., 2010). The  
10 summertime Arctic marine boundary layer (MBL) is left relatively clean after seasonal wet deposition of particles and reduced atmospheric transport of aerosols from anthropogenic sources at lower latitudes (Stohl, 2006; Browse et al., 2012; Croft et al., 2016). Such pristine conditions, combined with thermally stable MBL are typical of the Arctic summertime (e.g. Aliabadi et al., 2016). Clean Arctic air masses allow ultrafine (5 - 20 nm diameter) particle formation (Burkart et al., 2016), and the potential growth of secondary marine organic aerosols (including DMS-derived particles) into cloud condensation  
15 nuclei (Willis et al., 2016). Hence, the Arctic is a favourable terrain for new particle formation from biogenic DMS (Chang et al., 2011; Rempillo et al., 2011; Collins et al., 2017; Giamarelou et al., 2016; Mungall et al., 2016; Willis et al., 2016).”

**P3, L16:** “Could be particularly important” “In remote pristine marine areas such as the polar regions”.

**Response:** “Could be” and “Such as the polar regions” were added in the text (please see answer to P3, L11-15  
20 above).

**P3, L19:** Please add a reference here.

**Response:** References were added to the text (please see answer to P3, L11-15 above).

The sentence now reads: “[...] Such pristine conditions, combined with thermally stable MBL are typical of the Arctic  
25 summertime (e.g. Aliabadi et al., 2016). Clean Arctic air masses allow ultrafine (5 - 20 nm diameter) particle formation (Burkart et al., 2016), and the potential growth of secondary marine organic aerosols (including DMS-derived particles) into CCN (Willis et al., 2016).”

**P3, L20:** The study of Rempillo et al. (2011) could also be cited here.

**Response:** This reference was added to the text (see response to P3, L11-15 above).  
30

**P3, L22:** This statement is not true. Please read again Stefels et al. (2007). The 95% mentioned refer to the fraction of DMS emitted from the ocean, not to the fraction of DMS in natural reduced sulfur emissions. I think a few other references (e.g. Lana et al., 2011, or the work of Bates) might be more appropriate.

5 **Response:** We corrected this statement and added the suggested references. The new sentence reads: “Between 17.6 to 34.4 Tg of sulfur are released annually from the ocean to the atmosphere (Lana et al., 2011), accounting for 50-60% of the natural reduced sulfur emitted (Stefels et al., 2007).”.

**P3, L24:** The reference is not correct. It should be Green and Hatton (2014)

10 **Response:** The author’s names were corrected.

**P3, L23-24:** “Cellular metabolite” rather than “cellular compound”, compound is a little bit vague.

**Response:** We use the formulation “metabolite” as suggested.

15 **P3, L27:** I would suggest to cite Lyon et al. (2016) for the osmoregulation, especially since you are talking about phytoplankton and not algae. Similarly, Karsten et al. (1996) seems appropriate for the cryoprotection hypothesis.

**Response:** The suggested references were added in the text.

**P3, L32:** in-situ.

20 **Response:** “In situ” was written following BGD guidelines: “Common Latin phrases are not italicized (for example, et al., cf., e.g., a priori, **in situ**, bremsstrahlung, and eigenvalueound)”.

**P3, L33:** It would be nice to indicate in a short sentence how DMSP is released from the cell.

**Response:** Please see the additional information in the response to comment P3, L24.

25

**P3, L24:** Starting with “Between 1 and 40% of the DMSP...and ending page 3 line 9. The whole section is poorly structured and missing some important links. I would suggest to rewrite following these lines: “...found in several

phytoplankton species (DMSP particulate, or DMSP<sub>p</sub>) (see the review of Green and Hatton, 2014). DMSP plays several roles in phytoplankton, including osmoregulation (Lyon et al., 2016), cryoprotection (Karsten et al., 1996), and prevention of cellular oxidation (Sunda et al., 2002). Part of the DMSP produced by algae is released in the water column (dissolved DMSP, or DMSP<sub>d</sub>) where it is readily used by heterotrophic bacteria as carbon and sulfur sources (Kiene et al., 2000; Simó, 2001; Vila-Costa et al., 2006). The fraction of DMSP<sub>d</sub> consumed by heterotrophic bacteria and cleaved into DMS (DMS yield) may vary depending on the microbial community composition, its sulfur requirements, and the availability of other reduced forms of sulfur (Kiene et al., 2000; Stefels et al., 2007). DMSP-lyase enzymes are also present in several members of the microalgal groups Haptophyceae and Dinophyceae, and to a lesser extent Chrysophyceae (Niki et al., 2000). In addition to the DMSP cleavage pathway, a few studies have demonstrated the potential for reduction of dimethylsulfoxide (DMSO) by marine bacteria and phytoplankton (e.g. Spiess et al., 2009; Asher et al., 2011), and non-marine Antarctic shelf ponds bacteria (De Mora et al., 1996) as sources of DMS. This metabolic pathway is however not ubiquitous among bacterial assemblages (Hatton et al., 2004 ; Green and Hatton, 2014). DMS concentrations in surface mixed layers are further influenced by three sinks: bacterial and photo-oxidation to DMSO, and ventilation to the atmosphere (Bates et al., 1994; Kieber et al., 1996; Simó and Pedros-Alio, 1999b; del Valle et al. 2007, 2009). Two regimes of ocean DMS production are documented. A “bloom-driven” regime in eutrophic regions where the DMS concentrations are controlled by phytoplankton blooms (Stefels et al., 2007), and a “stress-driven” regime in oligotrophic open ocean regions, where DMS concentrations are highly correlated to UV radiation (Toole and Siegel, 2004), nutrient limitation (Stefels, 2000), in-situ –temperatures (Karsten et al., 1996; van Rijssel and Gieskes, 2002), and –salinity (e.g. Kirst, 1996). Ultimately, between 1 and 40% of the DMSP produced by algae reaches the atmosphere as DMS (Simó and Pedros-Alio, 1999a).”

20           **Response:** Sentences of the original paragraph were rearranged as suggested, with some modifications to facilitate the transition between paragraphs. The new proposed paragraph contains additional information regarding particulate DMSO (as recommended in specific comment P4, L5) and mechanisms for DMSP release from the cell (as recommended in specific comment P3, L330).

New paragraph: “DMS stems mainly from the enzymatic cleavage of dimethylsulfoniopropionate (DMSP) by algal and bacterial DMSP-lyases. DMSP is a cellular metabolite found in several phytoplankton species as particulate DMSP (DMSP<sub>p</sub>) (see the review of Green and Hatton, 2014). DMSP<sub>p</sub> plays various roles in phytoplankton, including osmoregulation (Lyon et al., 2016), cryoprotection (Karsten et al., 1996), and prevention of cellular oxidation (Sunda et al., 2002). Part of the DMSP<sub>p</sub> produced by algae is released in the water column as dissolved DMSP (DMSP<sub>d</sub>) via several pathways reviewed in Stefels et al. (2007), including active exudation, cell lysis, viral lysis and zooplankton grazing. DMSP<sub>d</sub> is then readily available to heterotrophic bacteria as carbon and sulfur sources (Kiene et al., 2000; Simo, 2001; Vila-Costa et al., 2006). The fraction of DMSP<sub>d</sub> consumed by heterotrophic bacteria and enzymatically cleaved by DMSP-lyases into DMS (DMS yield) may vary depending on the composition of microbial communities, their sulfur requirements, and the availability of other reduced forms of sulfur (Kiene et al., 2000; Stefels et al., 2007). DMSP-lyase enzymes are also present in several members of the

microalgal groups Haptophyceae and Dinophyceae, and to a lesser extent Chrysophyceae (Niki et al., 2000). Ultimately, between ~1 and 40% of the DMSP produced by algae reaches the atmosphere as DMS (Stefels et al., 2007; Simo and Pedros-Alio, 1999a).”.

5           **P4, L5:** It would be nice to write one or two sentences on particulate DMSO.

**Response:** The following sentences were added: “In addition to the DMSP enzymatic cleavage pathway, DMS production may arise from dimethylsulfoxide (DMSO) reduction by various groups of marine bacteria including proteobacteria (e.g. Vogt et al., 1997), members of the Roseobacter group (Gonzalez et al., 1999) and mat-forming cyanobacteria (van Bergeijk and Stall., 1996). However, the ubiquity of this DMSO-to-DMS reduction pathway amongst  
10 bacterial assemblages has not been established (Hatton et al., 2012). A limited number of phytoplankton species could also be involved in the reduction of DMSO into DMS (e.g. Fuse et al., 1995; Spiese et al., 2009). Increasing evidence suggests that particulate DMSO (DMSO<sub>p</sub>) may be directly synthesized by a potentially wide range of marine phytoplankton (e.g. Lee and de Mora, 1996) and could be involved in osmoprotection, cryoprotection (Lee and de Mora 1999), and anti-oxidant protective mechanisms (Sunda et al., 2002). As for dissolved DMSO (DMSO<sub>d</sub>), it is ubiquitous in seawater and continuous  
15 improvements in analytical techniques suggest that DMSO<sub>d</sub> may be as abundant as DMS in surface waters (e.g. Simo et al., 2000). DMSO is also a known sink for DMS (Hatton et al., 2004) via bacterial and photo oxidation of DMS to DMSO. Vertical Mixing and ventilation are also major removal processes influencing DMS concentrations in surface mixed layers (Bates et al., 1994; Kieber et al., 1996; Simó and Pedrós-Alió, 1999b; del Valle et al. 2007, 2009).”.

20           **P4, L10:** As explained in my general comments, I think you need here a paragraph on the importance of the sea ice ecosystem as a whole in the polar DMS cycle. This would help to better frame your study. It would be nice to introduce the important microbial biomass and DMS,P,O concentrations as well as the wide range of stresses encountered in the sea ice environment. Then you could talk about sea ice surface processes and introduce the cycling of DMS in melt-ponds. The review of Levasseur (2013) should help to put the melt-ponds in the general context of sea ice DMS production.

**Response:** A new paragraph was written as follows: “Ice-associated environments such as bottom sea ice, brine channels, melt ponds, under-ice surface waters, and leads provide complex and dynamic habitats to diverse microorganism communities involved in sulfur cycling (Levasseur, 2013). In the Arctic, the highest microalgal biomasses are found in the bottom ~0.1 m of sea ice, with Chlorophyll *a* (Chl *a*) concentrations several orders of magnitude above values for under-ice waters values (e.g. Legendre et al. 1992). A similar pattern of DMSP, DMSO and DMS build-up in bottom ice has been  
25 reported both in the Arctic and Antarctica (Kirst et al., 1991; Levasseur et al. 1994; Turner et al., 1995; DiTullio et al., 1995; Lee et al., 2001; Trevena et al., 2003; Trevena and Jones 2006; Delille et al., 2007; Tison et al., 2010; Asher et al., 2011; Nomura et al., 2012; Galindo et al., 2015). For example, DMSP<sub>p</sub> concentrations up to 15 000 nmol l<sup>-1</sup> have been documented  
30

during spring in bottom FYI of the Eastern Arctic (Galindo et al., 2014). DMSP, DMSO and DMS are also present throughout the ice column within the brine network (Levasseur et al., 1994; Trevena and Jones, 2006; Asher et al., 2011). Given that primary producers are the sole source of DMSP, very high ice concentrations of Chl *a* are often correlated with DMSP through a first order relationship (Levasseur, 2013). Beyond inter-specific differences in DMSP cellular contents (e.g. Keller et al., 1989; Stefels et al. 2004), environmental forcings are known to control DMSP, DMSO and DMS concentrations. In ice-associated environments, brine volume fraction might also be key in explaining DMS cycling variability via the control of ice permeability (Carnat et al., 2014). The melting season is a pivotal and productive period for these sulfur-containing compounds. Structural changes within sea ice during the melt season, namely increases in brine volume fraction and ice desalination, result in increased connectivity and permeability in the warming sea ice (Willis et al., 2006; Polashenski et al., 2012) and influence DMSP and DMS cycling (Carnat et al., 2014). Also, phytoplankton blooms developing under the ice during the melting period have been shown to produce large quantities of DMSP<sub>p</sub>, potentially leading to a build-up of DMS concentrations (Levasseur et al., 1994). In spite of the spatial importance of melt ponds, only few studies have investigated their role as a source of DMS for the Arctic atmosphere (e.g. Levasseur, 2013; Nomura et al., 2012).”.

15

**P4, L10-and further in the text.** There is also some DMS melt-pond concentrations in the study of Leck and Persson (1996). This study should be cited in your publication.

**Response:** Leck and Persson (1996) reference was added to the manuscript. The authors mention "negligible levels of DMS" in the "samples collected in the melt ponds [...]"encountered in "an area of multiple year ice".

20 The text was modified as follows: “Four studies have specifically reported on DMS in melt ponds so far. They reveal negligible DMS concentrations in MYI ice melt ponds in the Central Arctic Ocean, and concentrations up to 2.2 nmol l<sup>-1</sup> in the High Arctic (Sharma et al., 1999). In Antarctica, DMS concentrations ranging between 1.1 and 3.7 nmol l<sup>-1</sup> and between below the detection limit (d.l.) and 250 nmol l<sup>-1</sup> were measured in two studies (Nomura et al., 2012 and Asher et al. 2011, respectively).”.

25

**P4, L12:** Please check that the DMSO reduction mentioned by Asher et al. (2011) was effectively detected in melt-ponds. If I remember correctly, the experiment was made in brine rather than in melt-ponds. High DMSO and DMS concentrations were indeed observed in melt-ponds but I believe the tracer experiment was exclusively made in brine, which is a very different medium.

30 **Response:** Indeed, Asher et al. (2011) report results from experiments made in brine (as well as in slush and surface open waters), but not in melt ponds. The authors suggest that high DMS/P/O concentrations measured in melt ponds might



be associated with rapid DMS/P/O cycling and DMSO reduction, but future work is needed to firmly conclude. We clarified this in the amended version of the manuscript as follows: “In the latter study, bacterial DMSO reduction was suggested as a possible mechanism responsible for the high DMS concentrations observed although no actual rates of DMS production, either from DMSO or DMSP, were measured.”.

5

**P4, L10-15:** This is a little bit tricky. As you develop in the discussion section, the high DMS concentrations observed by Asher et al. (2011) were very likely related to the development of a surface ice community following flooding. I am fine with the fact that you develop this in the discussion section only, but I think you should already provide some hint in this introductory paragraph. It is a little bit misleading to only mention DMSO reduction and not to talk about the strong  
10 difference in microbial community development between the Arctic and Antarctic.

**Response:** The difference in surface communities between Arctic and Antarctic ecosystems, and the prevalence of surface ice communities following flooding was introduced as follows: “High DMS concentrations reported in the Antarctic are most likely related to the development of a surface ice community following flooding. Several studies document melt pond colonization by micro-, nano- and pico-sized algae as well as bacteria (Bursa, 1963; Gradinger et al., 2005; Elliott et al., 2015), suggesting that DMS in melt ponds may originate from algal and bacterial metabolism.”.  
15

Additional focus on the Antarctic versus Arctic sea ice dynamics are also provided in the discussion section (sect. 4.2.2). The text in the 4.4.2 section now states: “Extremely high gross DMS production rates from DMSO reduction, up to  $105 \pm 24$   $\text{nmol l}^{-1} \text{d}^{-1}$ , were measured within Antarctic sea ice brines by Asher et al. (2011). The authors suggested that this mechanism could also potentially be responsible for the high DMS concentrations (up to  $250 \text{ nmol l}^{-1}$ ) measured in Antarctic melt ponds. The absence of DMS production from  $^{13}\text{C}$ -DMSO in the melt ponds studied here may then reflect potential differences in microbial assemblages within melt ponds, as the metabolic ability to convert DMSO into DMS is not ubiquitous among bacterial communities (Hatton et al., 2012; Hughes et al., 2014). In support of this hypothesis, it has been shown that between 70 and 78% of the operational taxonomic units (OTU), a marker of microbial diversity, in Arctic and Southern Ocean surface water communities are unique to their region (Ghiglione et al., 2012). Observed differences in the  
20 biological characteristics of melt ponds between the poles could also reflect divergent sea ice dynamics. Antarctic sea ice salinity is higher by 0.5 to 1.0% than in Arctic sea ice (Gow et al., 1982, 1987) and the C-shaped salinity profile that is typical in fully formed Arctic FYI is not as prominent in Southern Ocean sea ice (Eicken, 1992). Antarctic sea ice is commonly subjected to intense rafting. Flooding, a process whereby heavy snow load pushes the ice below the water level, is common in the Antarctic and results in the formation of snow ice (Hunke et al., 2011). Antarctic melt ponds studied in Asher et al. (2011) may have been subjected to this flooding leading to the formation of salted “freeboard layers” (Haas et al.,  
25 30 et al., 2001; Massom et al., 2006). This is supported by the reported highest salinities in the top sea ice layers and the subsequent

salinity decrease throughout the ice profile (Asher et al. 2011). Such configuration may bring highly productive microbial communities at the surface of the ice, potentially responsible for the high DMS concentrations observed in melt ponds. [...].”

5 **P4, L15:** “may also originate”. Remove the also. You did not provide another explanation for the presence of DMS in the Arctic melt-ponds so far in the text.

**Response:** The word “also” was removed.

**P4, L16:** It would great to include here a few sentences on the typical environmental conditions/stress developing in surface melt-ponds, and how these conditions could influence DMS(P) production.

10 **Response:** We agree that introducing the incidence of known plankton stressors such as elevated light, substrate limitation and osmotic shock would make valuable additions. However, the length of the introduction has already considerably increased in the revised version of the manuscript. We therefore decided to not add this additional information to the introduction. We mention the typical environmental stress in melt ponds in several instances throughout the manuscript (e.g. In the introduction: “environmental forcings are known to control DMSP, DMSO and DMS concentrations”  
15 and in section 4.2.3 “Fast and transient intracellular accumulation of compatible solutes, such as DMSP, may serve as an adaptive strategy by microbial cells to help cope with fluctuations of the surrounding environment, increasing their tolerance to osmotic and thermal stresses for example (Welsh, 2000).”).

20 **P4, L17-18:** It would be nice to rephrase and develop a little bit more this paragraph. The reader must be able to clearly identify the questions/gaps your study is going to address. For now it reads like the paper is just another data report ...while I believe it is much more than that. Make it a little bit more appealing.

**Response:** The rewritten paragraph reads as follow:

25 “Considerable efforts have been dedicated to the understanding of underlying process controlling the physics of melt ponds and their feedbacks on climate through the control of surface energy balance of the ice (Lüthje et al., 2006; Polashenski et al., 2017). However, little is known about their biogeochemistry. Four studies have specifically reported on DMS in melt ponds so far. They reveal negligible DMS concentrations in MYI ice melt ponds in the Central Arctic Ocean, and concentrations up to 2.2 nmol l<sup>-1</sup> in the High Arctic (Sharma et al., 1999). In Antarctica, DMS concentrations ranging between 1.1 and 3.7 nmol l<sup>-1</sup> and between below the detection limit (d.l.) and 250 nmol l<sup>-1</sup> were measured in two studies (Nomura et al., 2012 and Asher et al. 2011, respectively). In the latter study, bacterial DMSO reduction was suggested as a  
30 possible mechanism responsible for the high DMS concentrations observed although no actual rates of DMS production,

either from DMSO or DMSP, were measured. High DMS concentrations reported in the Antarctic are most likely related to the development of a surface ice community following flooding. Several studies document melt pond colonization by micro-, nano- and pico-sized algae as well as bacteria (Bursa, 1963; Gradinger et al., 2005; Elliott et al., 2015), suggesting that DMS in melt ponds may originate from algal and bacterial metabolism. Yet, in situ DMS production had never been measured nor had key mechanisms been identified. Here, we report on the DMS concentrations in nine melt ponds located in the Eastern Canadian Arctic Archipelago (CAA), and on the prerequisites and processes responsible for the presence of this climate-active gas. This is the first attempt to assess the dynamics of DMS in Arctic melt ponds. We identified sea ice permeability as a major control of DMS production in melt ponds, mediating the transport of both DMS and DMS-producing communities toward the surface of sea ice. We also provide the first evidence for direct in situ DMS production in Arctic melt ponds. We propose that seasonally melting sea ice might become increasingly prone to DMS production as FYI become largely predominant at the regional scale.”

**P4, L25:** You could already indicate here between brackets (logistical constraints) why basic physical measurements were not conducted at Ice2.

**Response:** “(logistical constraints associated with the ship time line)” was added in the first sentence of the paragraph.

**P4, L26:** Please already define freeboard here.

**Response:** We added a definition of freeboard in the text: “[...] freeboard (the height of sea ice above the ocean surface), [...]”.

**P4, L26:** What motivated the sampling at a 3 m distance? Did you collect any other cores than the ones mentioned in this study? It would be nice to have an idea of the ice/snow thickness variability around the melt-ponds sampled.

**Response:** The text now states : “The 3 m distance was a compromise between maximizing the proximity of ice and melt pond samples and minimizing melt pond disturbance during sampling operations. Ice and freeboard thickness presented in table 1 are averaged values of the 7 (Ice1) to 8 (Ice3 and Ice4) ice cores sampled at each station between the team members for their respective projects.” We aslo modified the caption accordingly: “Table 1: Physical characteristics of the sea ice surrounding the melt ponds. Note that only melt pond sampling (i.e. no ice sampling) was conducted at station Ice2 due to ship-related logistical constraints. A negative freeboard height indicates that the ice surface was locally below the mean sea level. n/a stands for non-available data. Ice thickness and freeboard values are averages of 7 (Ice1) to 8 (Ice3 and Ice4) ice cores sampled at each station.”

**P4, L27:** For sea ice physics discussions, it is always easier to measure salinity and temperature on the same ice core and at the same vertical resolution. It is always better to make full depth profiles as you will see later in my comments.

**Response:** Full ice depth temperatures and salinity profiles are now presented in a figure (new figure 3) and included in the discussion. Please see response to comment P 12, L27 – P13, L14 for further details.

**P4, L28-29:** Remove “According to a widely used protocol” and all the references that follow. Write: Sea ice temperature and bulk ice salinity were measured following Miller et al. (2015). Then: “Sea ice temperature was ...”.

**Response:** The phrase ‘According to a widely...’ was removed and the reference Miller et al (2015) was added.

10

**P4, L30:** (and throughout the manuscript). Check for spacing between 5 and cm. I do not know what the recommendations of Biogeosciences are.

**Response:** Biogeosciences recommends “not to hyphenate modifiers containing abbreviated units (e.g. "3-m stick" should be "3 m stick")”. As we did not find any other recommendations, we inserted non-breaking spaces throughout the manuscript to avoid line breaking between numbers and their units.

15

**P4, L28-31:** Precision/accuracy of the probes should be indicated when available. Also check if you need to add trademark symbols next to the brands.

**Response:** Precision of the probes were indicated in the amended manuscript. Trademarks/registered symbols were revised throughout the text.

20

**P4, L32:** “the bulk salinity of the melt aliquot”.

**Response:** As suggested by the Anonymous Referee #1, we changed this to “Bulk salinity of the melted ice section”.

25

**P4, L32:** Permeability to fluid/gas transport is a more appropriate term than porosity here.

**Response:** The sentence was changed to: “Permeability to fluid transport was assessed with brine volume profile calculations from bulk salinities and sea ice temperatures following equations from Leppäranta and Manninen (1988) for sea ice temperatures  $> -2^{\circ}\text{C}$  (Fig. 3).”.

5           **P5, L1-3:** and further in the discussion. Here you need to calculate the brine volume fraction in your sea ice samples following Leppäranta and Manninen (1988). The section needs to be rewritten. You cannot talk about permeability/porosity and the rule of fives without calculating and using the brine volume fraction. The rule of fives refers to three fives, salinity, temperature, and over all brine volume fraction. Temperature and salinity only are not sufficient to discuss permeability issues. Golden’s research and all the research conducted on sea ice permeability and its influence on  
10 biogeochemistry (see Carnat et al. (2013), Carnat et al. (2014), Jardon et al. (2013), Zhou et al. (2013) indicate that sea ice becomes permeable to fluid transport when brine volume fraction reaches 5% (note that this threshold might vary substantially depending on ice texture for instance). The rule of fives stipulates that such a brine volume fraction (5%) corresponds for instance to a temperature of  $-5^{\circ}\text{C}$  for an ice salinity of 5...not that the ice is permeable when the ice temperature is warmer than  $-5^{\circ}\text{C}$  and the salinity higher than 5.

15           **Response:** Full ice depth T and S profiles are now presented in a new figure (Fig. 3) for stations Ice1, Ice3 and Ice4. Corresponding brine volume profiles were calculated using the recorded sea ice bulk salinity and in situ temperature (Leppäranta and Manninen, 1988). Calculated brine volume fraction constantly exceeded 10% in the ice profiles, except in the upper 0.1 m section of the Ice3 station. For the latter, we likely observed the effects of refreezing metamorphosis of snow and / or sea ice recrystallization. As mentioned in Polashenski et al. (2017) after Golden et al. (1998) and Golden (2003),  
20 liquid inclusions in columnar sea ice become interconnected once brine volume fraction reaches 5% in columnar FYI and 10% in granular FYI. Although no ice structure analysis was conducted during our study, columnar ice is expected to dominate FYI stratigraphy in the Arctic (Thomas and Dieckmann, 2008). We therefore decided to use the 5% brine volume threshold for the ice permeability in our study. These additional data show that the sea ice was (with the exception of the upper 0.1 m in Ice3 ice core) highly permeable throughout the three full ice profiles.

25

**P5, L7:** Additional details are needed here. It is not clear to me what the maximum pond fraction is. A picture of melt ponds has one and only one melt pond fraction. Regarding the mean, did you calculate it from multiple pictures? Could you provide the approximate area covered by the pictures? How many pictures were taken for each site? Did you try to assess the pond coverage digitally? Perhaps it would be great to indicate your estimated pond fraction for each sampling  
30 location in Fig1.

**Response:** We did not assess the melt pond fraction (MPF) digitally. Although we agree that a digital assessment of MPF would represent valuable information, detection and quantification of sea ice surfaces, including melt ponds coverage,

is still an ongoing research field (Scharien et al., 2017 ; Wright and Polashenski, submitted). Impact of melt ponds on the ice albedo (e.g. Flocco et al., 2012), and the link between spring melt pond fraction and September sea ice minimum extent (Schröder et al., 2014), both using MPF, are still actively explored. Various techniques including low-level aerial photographs (e.g. Derksen et al., 1997), satellite based passive microwave observations (Fetterer and Untersteiner, 1998),  
5 synthetic aperture radar (Yackel and Barber, 2000), Moderate Resolution Imaging Spectroradiometer imagery (MODIS) (e.g. Tschudi et al., 2008) and LiDAR (Light Detection and Ranging) (Landy et al., 2014) are used throughout the literature. These techniques are beyond the scope of the present study. In addition, MODIS data of melt pond fraction after 2011 are not publicly available yet. Also, the use of melt pond coverage data is minimal in our study. That being said, the term “maximum” was removed from the text as it infers that there was also a minimum pond fraction at each station. Between two  
10 and three persons documented the sampling operations by taking 5 to 10 digital photographs using a hand-held camera from the bridge (17 m height) during each station. Their individual assessments were then compared and averaged values are presented. An approximative size scale and the estimated MPF originally presented in Table 1 were added to figure 1 and removed from Table 1.

15 **P5, L11:** How many replicates? It is not clear if chl *a* was measured on the ship or the filters stored.

**Response:** Measurements were done in duplicate. This was changed in the manuscript. Chl *a* measurements were conducted on board. The text now states: “[...] duplicates of in situ pond water were filtered onto Whatman GF/F 25 mm filters. Pigments were extracted in 90% acetone for 18 to 24 h in the dark at 4°C (Parsons et al., 1984). Fluorescence of the extracted pigments was measured on board with a 10-005R Turner Designs fluorometer [...]”.

20

**P5, L23-24:** This is slightly confusing. Stored in liquid nitrogen (-196°C) or kept frozen at -80°C?

**Response:** Duplicate 4 ml subsamples were fixed with 20 µl of 25% glutaraldehyde Grade I (0.1% final concentration; Sigma-Aldrich G5882), then subjected to quick-freeze in liquid nitrogen for 24h, and finally stored at -80°C until analysis. This is now added in the manuscript.

25

**P5, L27:** Did you consider sampling multiple depths in the melt-ponds? Would you have expected homogeneity or a vertical gradient? Please quickly discuss this in the text.

**Response:** Strong saline stratification can be found in Antarctic (terrestrial) ponds but seems to be specific to deep ponds (0.5-1.5 m) (Wait et al., 2006). Jung et al. (2015) also report highly stratified open melt ponds (i.e. melt ponds that  
30 have melted all the way to the sea surface) in Arctic FYI. However, closed FYI melt pond modelling suggests that convective- and wind-driven- mixing generate well-mixed melt ponds and stratification is not a significant factor in melt

pond circulation (Skylvingstad and Paulson, 2007). Some temporary stratification might be expected in the melt ponds in the absence of wind but this is rapidly (a few hours) overturned by solar heating-driven convection. The following information was added in the paragraph:

5 “Stratified open melt ponds (i.e. melt ponds that have melted all the way to the sea surface) were reported in Arctic FYI (Jung et al., 2015). However, closed FYI melt pond, such as those sampled during this study, are not prone to vertical stratification due to convective- and wind-driven- mixing (Skylvingstad and Paulson, 2007). Given their shallow depths (less than 0.3 m), melt pond stratification was most probably inexistent or minimal during our study.”.

**P5, L30:** “to fill the glass serum bottles” remove the “the”.

10 **Response:** “the” was removed

**P6, L11:** Consider cutting in two sentences. “...into 5 ml Falcon™ tube. DMSPd was quantified ...”.

**Response:** Sentence was cut in two as suggested.

15 **P6, L13:** Please provide whenever possible an estimate of the error associated with every measurement. This is clearly missing for the measurement of DMS(P) concentrations.

**Response:** Measurement error estimates were added to the text.

**P6, L16:** Dacey and Blough (1987) is perhaps a better reference here than Levasseur et al. (2006).

20 **Response:** The reference was changed in the text as suggested.

**P6, L26:** “freshwater”, do you mean milliQ water? Please specify.

**Response:** Yes, milliQ water is now specified in the text.

25 **P6, L30:** Consider using “duplicate” instead of “duplicated”.

**Response:** “Duplicated” was changed to “duplicate”.

**P7, L10:** This is I think the first time a Table is mentioned in the text. It should then be Table 1. I suggest to add a reference to Table 1 earlier in the text, in section 2.1.

**Response:** We kept the original Table 1 but mention it earlier, in section 2.1. (Table 1 “ Table 1: Physical characteristics of the sea ice surrounding the melt ponds. [...].) The first mention of Table 2 is lower in the text, in the paragraph preceding section 2.2.

**P8, L5:** Is any fractionation expected during storage?

**Response:** Isotopes fractionation may be caused by differences in rates of reaction or diffusion, or by differences in equilibrium constants. Fractionation during prolonged (several months) storage has been noted before in nitrogen cycle studies, even for frozen samples (Thayer, 1970; Granger et al., 2006). According to kinetics theory, kinetic energy (K.E.) is the same for all gases at a given temperature, which can result in greater velocities of lighter isotopes compared with their heavier counterparts (Sharp, 2007). Detailed calculation is provided in the Supplements to the response to referee #2 in the interactive comments section of Biogeosciences Discussion. We find that average velocity of DMS (m/z 62) is 0.8% greater than the average velocity of DMS (m/z 63) molecules in the same system. Following the same calculation steps, average velocity of DMS (m/z 62) is 4.7% greater than the average velocity of DMS (m/z 68) molecules in the same system. Finally, average velocity of DMS (m/z 63) is 3.8% greater than the average velocity of DMS (m/z 68) molecules in the same system. Accordingly, a negligible maximum fractionation of 5% is expected during storage. Preserved samples and standards were compared against standard curves and fractionation during storage was not observed.

**P8, L6-10:** Please provide the overall precision of the methods.

**Response:** The precision of the method was provided.

**P9, L5:** Please add this 5 m information in the section 2.1 of the materials and methods part.

**Response:** The distance information was added to the method section.

**P9, L6-8:** Following my previous comments, this section needs to be rewritten. Also refrozen snow at the surface means superimposed ice, an ice texture known to be impermeable. This should be mention somewhere in the text.

**Response:** Both reviewers pointed out the shortcomings of the section dealing with ice physics. To address this, full ice depth temperature and salinity profiles are now presented. Corresponding brine volume fraction were calculated, and



presented alongside with salinity and temperature profiles in an additional figure (see new figure 3). Calculated brine volume fractions are now used to discuss sea ice permeability. The method section was also amended accordingly to reflect this additional dataset. We also mention that the refrozen snow observed at station Ice3 was impermeable and may be indicative of refreezing metamorphosis of snow.

5

**P9, L29:** Please replace (see discussion) by “This will be discussed in section...”.

**Response:** The text was modified to “This will be discussed in sect. 4.2.2.”.

**P11, L11:** The use of “significantly” implies a statistical test which is not provided.

10 **Response:** Statistical test is now provided in the text as follows: “During both Ice1-MP1 and Ice4-MP1 incubation experiments, the Light versus Dark Treatment had no effect on the net changes in DMSP<sub>d</sub> concentrations between the L-DMSP/O and D-DMSP/O Treatments (Wilcoxon Signed-rank test; n=8, df=3,  $\alpha=0.05$ ), but significantly impacted the rates of net accumulation of DMS (Wilcoxon Signed-rank test; n=12, df=5,  $\alpha=0.05$ ). The accumulation of DMS over 24h in the L-DMSP/O Treatments were consistently and significantly lower than in the corresponding D-DMSP/O Treatments (Wilcoxon  
15 Signed-rank test; n=8, df=3,  $\alpha=0.05$ ).”.

**P11, L17-30:** You could make the paragraph a little bit lighter to read and easier to follow by removing some unnecessary instances of (m/z 68) and (m/z 62).

**Response:** As suggested, several mentions of “m/z” have been removed in the paragraph.

20

**P12, L1-2:** See my previous comment. Please read the study of Leck and Persson (1996), cited in Levasseur (2013). There is also some interesting work in glacial melt water ponds that you could consult and perhaps cite somewhere in the manuscript (De Mora et al., 1996), especially regarding to DMSO as a source of DMS.

25 **Response:** Studies by Leck and Persson (1996), Sharma et al. (1999), Nomura et al. (2012), and Asher et al. (2011) are now cited regarding DMS concentrations in sea ice melt ponds. De Mora et al. (1996) is cited in the introduction.

**P12, L5:** As stated before, I think this sentence is misleading and should be removed giving the fact that you provide further in the text a very plausible explanation for the difference. This explanation is moreover relatively logic for someone with a basic knowledge of sea ice biogeochemistry.

25

**Response:** The sentence stating that “Our current limited understanding of the mechanisms responsible for the cycling of DMS in melt ponds prevents the identification of the underlying causes of these differences” was removed from the manuscript.

5           **P12, L14:** What do you mean by “closed melt pond”? It seems that the melt-pond is exchanging material with seawater and the atmosphere. Please clarify.

**Response:** Closed melt pond terminology refers to “closed bottom” light blue coloured melt ponds that form on relatively thick ice cover. This is the only type of melt ponds discussed in our study. Open melt ponds on the contrary are dark blue ponds directly connected to the underlying seawater with a visible hole in the relatively thin ice cover. This  
10 terminology was borrowed from Lee et al., 2012.

**P12, L17 and 23:** “Sea spray”.

**Response:** The typo was corrected.

15           **P12, L27 – P13, L14:** This whole section needs some rewriting. Full-depth gravity drainage should not be confused with flushing of surface melt-water. You should read a little bit more carefully the study of Jardon et al. (2013), but also Carnat et al. (2013) which describes the seasonal evolution of sea ice salinity (and brine salinity) in FYI in the Canadian Arctic (Amundsen Gulf, Beaufort Sea)[...].

**Response:** Comment on paragraph P12, L27 – P13, L14 was extensive and called for a re-writing of the full  
20 paragraph so we respond to each point separately in the following section:

          Proper terminology (brine flushing versus brine drainage) is used in the corrected version. As discussed in the response to referee #1 (P13, L10), calculations provided by Jardon et al. (2013) deal with the permeability threshold of sea ice with salinity greater than 5. With a bulk sea ice salinity of 2.79 (averaged for the three stations), and a maximum value of 5.00 at station ice4 (1.2-1.3 m section), our melt ponds fall outside of this range. The statement regarding full ice depth  
25 desalination referencing Jardon et al. (2013) was removed from the manuscript.

          [...] Also, you definitely need to include brine volume fraction, Rayleigh number, and brine salinity here in the discussion. Unfortunately you only measured surface ice salinity and temperature, while full-depth profiles are generally necessary for this type of discussion. For instance, you could have 10 cm of sea ice with a low salinity due to percolating melt water with more saline layers underneath. Full-depth gravity drainage/convection requires both a connected brine

network (sea ice permeable to fluid transport), and hence usually brine volumes above 5%, and an unstable brine density (brine salinity) profile [...].

**Response:** We took full ice depth temperature and salinity profiles during the study. We chose to only use the upper 0.1 m measurements in sea ice in the submitted version of the manuscript to illustrate the physical conditions of the ice closest to the melt ponds. Given that we discuss ice physics and permeability, we agree that it is necessary to include the full ice profiles (salinity, temperature, and brine volume) in the revised version. These results are now presented in a new figure 3 and discussed in the revised manuscript. Averaged values for bulk sea ice salinity over the full thickness of the ice were 1.73, 2.83 and 3.75 at stations Ice1, Ice3 and Ice4, respectively. Locally, maximum bulk salinity never exceeded 5.00 (Ice4, 1.2-1.3 m section). In situ temperatures, averaged over the full thickness of the ice, were -0.54 °C, -0.52 °C and -0.98 °C at stations Ice1, Ice3 and Ice4, respectively, and reached a minimum value of -1.39 °C (Ice4, 0.8-0.9 m section). Brine volume fraction constantly exceeded 10% in the ice profiles, except in the upper 0.1 m section of the Ice3 station where we likely observed the effects of refreezing metamorphosis of snow and / or sea ice recrystallization (as mentioned in the response to P5, L1-3).

[...] The combination of these two criteria can be expressed via a Rayleigh number. When sea ice warms up and reach the permeability threshold (expressed by the brine volume fraction, not the temperature), instability of the brine network (brine salinity being a direct function of sea ice temperature (Cox and Weeks (1983)), colder surface ice has saltier and denser brine than warmer bottom ice) can result in full-depth convection, brine being replaced by upward moving seawater. This usually occurs in mid-late spring (see the study of Carnat et al. (2013)) and results in some desalination of the ice cover (the upward moving seawater being less saline than the brine it is replacing). Following further warming in summer, surface melt water (melting snow or melting surface sea ice) percolates within the brine net work leading to the process called flushing. This further decreases the bulk ice salinity down to values way under 2 as observed in your study. Warming will also dilute brine with pure ice melt water. I think that at the time of your sampling (based on the limited salinity and temperature data available), both full-depth gravity drainage and some flushing have already occurred. Hence, brine cannot indeed be responsible for the salinity observed in the melt-ponds. [...].

**Response:** Because of the apparent advanced desalination of sea ice in the ice cores presented, we did not include the Rayleigh (Ra) number results. We present the detailed calculation in the supplement to the response to referee #2 in the interactive comments section of Biogeosciences Discussion. Because of the high in situ ice temperatures and the low brine salinities, two terms used in Ra computation, we found negative values of Ra. Given that errors in Ra are largest for warm and permeable sea ice (Vancoppenolle et al., 2013), we decided to exclude these calculations in the reviewed version of the paper. With winter gravity drainage, flushing is the dominant desalination process for fully formed sea ice. Flushing is the three dimensional (i.e. both vertical and laterally in all directions) washing out of salty brine from the structure of porous sea ice and its replacement with a mix of seawater and melt water (Hunke et al., 2011). With our averaged bulk salinity < 4.00

throughout the ice, we agree that sea ice had most likely undergone full-depth salinity drainage and brine flushing before our sampling.

5 New paragraph: “Ice brine intrusion is also unlikely to have contributed significantly to melt pond salinization since the averaged bulk ice salinity was low (under 5), especially in the top 0.2 m where it did not exceeded 2. It is also known that most of the hyper-saline brine characterizing consolidated cold FYI in winter are lost in spring through full depth brine convection well before melt ponds start to form (Jardon et al., 2013). Residual salts are finally lost through meltwater flushing during the summer season (Weeks and Ackley, 1986, Eicken et al., 2002; Vancoppenolle et al., 2007). At the time of our sampling, low bulk salinity values, combined with calculated brine volume fraction constantly exceeding 10% in the entire sea ice profiles (except in the upper 0.1 m section of the Ice3 station) suggest that full depth flushing had already  
10 occurred. We thus exclude sea ice brine enrichment of melt ponds as their main salinization mechanism.”.

[...] Now you still have to explain how to get seawater in contact with the melt-pond water through the porous brine network. Full-depth gravity drainage as suggested P13L10 makes no sense to me as the brine salinity do no support instability anymore. You also have to be a little bit careful with the use of the freeboard, especially citing Hudier et al. (1995). What Hudier et al. (1995) refers to is the loading of the sea ice surface with a significant amount of snow, depressing  
15 the surface sea ice level below the seawater level, leading to flooding of the ice surface, followed by gravity drainage. This is not really what you observed here. I agree that the decrease in sea ice thickness and development of the melt pond translate into a loss of freeboard, and that the melt-pond depth might approach the freeboard height, or even get below that height. Given the height of the freeboard and the depth of the melt-pond, seawater might infiltrate the porous ice texture via the brine network and start exchanging with the melt-pond. I am a little puzzled by the diffusion mechanism you suggest. It is  
20 probably true that at some point of the melt-pond evolution, infiltrated melt water might freeze and block the flushing of the pond by decreasing permeability in the ice layer under the melt-pond. No direct exchange with underlying seawater would then be possible. Diffusion could occur but would be a very slow process (especially through such layer), rather unlikely to explain the salinity change and biomass seeding observed in the pond. Alternatively, I wonder if the pond evolution could not alternate between phases of flushing, and phases of replenishment (pond depth being close to or below the freeboard  
25 height) with a mix of seawater and pond water. These phases would be controlled by small changes in ice temperature oscillating around the freezing temperature of the melt water. I think that the similarity in species composition between the melt-pond and under-ice seawater supports well this mechanism.

**Response:** The paragraph was modified as follows: “Rather, we suggest that melt ponds salinization originated mostly from the intrusion of seawater through the ice. Although closed melt ponds are not visibly connected to seawater,  
30 exchanges with the underlying seawater can take place. The extent of these exchanges are dependent on the sea ice freeboard and micro-structure, i.e. the amount, size and shape of brine inclusions (Carnat et al., 2014), that controls sea ice permeability. Above a critical brine volume ranging between 5% (for columnar sea ice) and 10% (for granular sea ice), brine inclusions become interconnected. During the melting season, decrease in sea ice thickness is enhanced by the formation of

the melt pond and lead to a loss of freeboard. As melt ponds become closely levelled with seawater, small changes in ice temperature oscillating around the freezing temperature may result in episodic intrusion of seawater mixed with meltwater through the porous ice. Seawater mixed with meltwater penetrating the brines channels of permeable sea ice may bring salts, nutrients and microorganisms (Jardon et al., 2013, Vancoppenolle et al., 2010), potentially reaching surface melt ponds. This mechanism most probably explains the salinity and biochemical characteristics of Ice1 and Ice4 melt ponds. Station Ice3 represents a different case. Here, the low melt pond salinity (and absence of biological activity) may be explained by the presence of an impermeable ice layer on the top of the ice preventing both pond drainage and exchange between pond water and seawater.

We also considered the following paragraph as a valuable addition to the discussion: “We acknowledge that our data set is too limited to draw firm conclusions on the processes governing the formation and salinization of FYI melt ponds. Yet, in the interest of further research, we conjecture that snow load before melt onset may be crucial in determining the fate of melt ponds not only with regards to their saline status, but also their potential to produce DMS. Brine volume, derived from bulk salinity and temperature, generally provides a valid proxy for sea ice permeability. In some case however, melting of high snowpack generates a considerable flow (up to 15cm d<sup>-1</sup>) of freshwater into the porous structure of sea ice (Polashenski et al., 2017). This can create localized ice plugs within the highly connected brine network of apparently porous sea ice and allow melt ponds to persist above sea level well after sea ice bulk sea ice brine volume reached a critical level (5-10%). Such deviation from the porosity/permeability relationship following freshwater intrusion is demonstrated in Polashenski et al. (2017). We suggest that we observed such case of melt pond persistence above sea level in station Ice3. Alternatively, lower snow load remaining at the onset of the melt season will translate into a less abundant freshwater input above sea ice. Snow load distribution is however notoriously highly variable even at the meter scale due to wind redistribution and sea ice topography variability (e.g. Polashenski et al., 2017). Low snowpack would induce limited insulation of the sea ice from atmospheric conditions, resulting in 1) a more gradual warming of sea ice during spring season, and 2) limited freshwater loading available for percolation blockage. In such case, freshwater would not seal the ice through percolation blockage (Polashenski et al., 2017). Sea ice would then remain entirely porous as soon as the 5-10% brine volume threshold is reached, facilitating melt pond salinization process. We suggest that this scenario may have been observed at stations Ice1 and Ice4.”

**P14, L6-8:** “over-flooding of sea ice”. Replace by “flooding of the ice surface”. Over-flooding is an odd term.

**Response:** “Over-flooding” is no longer used throughout the text.

**P14, L6:** Flooding could be better defined.

**Response:** Flooding was redefined in the introduction as follows: “[...] flooding, a process whereby heavy snow load pushes the ice below the water level. Flooding is common in the Antarctic and results in the formation of snow ice (Hunke et al., 2011).”.

5 **P16, L7:** Again, consider other data sets available.

**Response:** Datasets from Nomura et al. (2012) and Leck and Persson (1996) are now considered.

**P16, L16:** Modify “over-flooding”.

**Response:** Over-flooding was modified to flooding as suggested in P14, L6-8.

10

**P16, L20:** There are several studies providing direct (Nomura et al., (2012) and indirect (Carnat et al., (2014)) evidences of DMS flux from FYI surface toward the atmosphere.

15 **Response:** These references were added to the manuscript. The text now reads: “To this day, most climatologies assume the absence of DMS fluxes above ice-covered waters (e.g. Lana et al., 2011) even though several studies provide direct (Zemmelink et al., 2008; Nomura et al., 2012, MYI) and indirect (Carnat et al.,2014, FYI) evidence of DMS venting from snow-covered Antarctic sea ice.”.

**P16, L24:** These numbers should be put in perspective. How do they compare to the sea ice, ocean reservoirs?

20 **Response:** As explained previously (response to comment P2, L11-15), we deleted the estimate of the size of DMS reservoir in Arctic FYI melt ponds from the manuscript.

**P16, L26:** Is the average depth calculated from your data set or from literature observations? Your data set is relatively small.

25 **Response:** This is the average depth for the 9 melt ponds measured during this study. We agree that a greater spatial coverage of melt ponds is needed to come up with a more robust estimate and we therefore deleted the section of the discussion (and abstract) where we tentatively estimated the contribution of melt ponds to the overall size of the DMS reservoir in Arctic.

**P16, L29:** Wind velocity but also a better understanding of gas exchange between small fetch melt ponds and the atmosphere.

**Response:** The necessity of a better understanding of gas exchange between small fetch melt ponds and the atmosphere was added to the manuscript.

5

**References:** Check the alphabetic order, Giamarelou et al. should be after Garrison.

**Response:** The alphabetical order of references was checked and corrected.

### Figures and tables

10

**Table 2:** check the significant digits in the temperature values. Only physical characteristics are presented here, remove the chemical and biological characteristics from the caption.

**Response:** Temperature values are now expressed with only one significant digit in Table 2 and the title of the table was corrected as suggested.

15

**Table 7:** Please be consistent with the significant digits.

**Response:** Done.

20

**Figure 1:** Please add a scale on figure 1b. As requested above, it would be nice to indicate the melt-pond fractions on each picture and an explanation of the calculation in the caption.

**Response:** Scales were added on figures 1b and 1c along an estimate of pond fraction.

**Figure 3:** Odd lettering of the figures.

**Response:** (\*Now figure 4) The numbering was modified so that “c” and “b” are now interchanged.

25

# Marked-up manuscript version of: Dimethylsulfide dynamics in first-year sea ice melt ponds in the Canadian Arctic Archipelago

Margaux Gourdal<sup>1</sup>, Martine Lizotte<sup>1</sup>, Guillaume Massé<sup>1</sup>, Michel Gosselin<sup>2</sup>, Michel Poulin<sup>4</sup>, Michael Scarratt<sup>3</sup>, Joannie Charette<sup>2</sup>, Maurice Levasseur<sup>1</sup>

<sup>1</sup>Département de biologie, Québec-Océan and Unité Mixte Internationale 3376 TAKUVIK, CNRS-Université Laval, 1045 avenue de la Médecine, Québec, Québec G1V 0A6, Canada

<sup>2</sup>Institut des Sciences de la Mer de Rimouski (ISMER), Université du Québec à Rimouski, 310 allée des Ursulines, Rimouski, Québec G5L 3A1, Canada

10 <sup>3</sup>Maurice Lamontagne Institute, Fisheries and Oceans Canada, P.O. Box 1000, Mont-Joli, Québec G5H 3Z4, Canada

<sup>4</sup>Research and Collections, Canadian Museum of Nature, P.O. Box 3443, Station D, Ottawa, Ontario K1P 6P4, Canada

*Correspondence to:* Margaux Gourdal (margaux.gourdal@takuvik.ulaval.ca)

## Copyright statement

© Author(s) 2017. CC-BY 3.0 License.

15

Changes tracking colour code:

Changes suggested by anonymous referee #1 – Changes suggested by referee #2 – Authors additions

20



## Abstract.

Melt pond formation is a natural seasonal pan-Arctic process. During the thawing season, melt ponds may cover up to 90% of the Arctic first-year sea ice (FYI), and 15 to 25% of the multi-year sea ice (MYI). These pools of water lying at the surface of the sea ice cover are habitats for microorganisms and represent a potential source of the biogenic gas dimethylsulfide (DMS) for the atmosphere. Here we report on the concentrations and dynamics of DMS in nine melt ponds sampled in July 2014 in the Eastern Canadian Arctic Archipelago. DMS concentrations were under the detection limit ( $< 0.01 \text{ nmol l}^{-1}$ ) in freshwater melt ponds, and increased linearly with salinity ( $r_s = 0.84$ ,  $p \leq 0.05$ ) from  $\sim 3$  up to  $\sim 6 \text{ nmol l}^{-1}$  (avg.  $3.7 \pm 1.6 \text{ nmol l}^{-1}$ ) in brackish melt pond. This relationship suggests that the intrusion of seawater in melt ponds is a key physical mechanism responsible for the presence of DMS. Experiments were conducted with water from three melt ponds incubated for 24h with and without the addition of two stable isotope-labelled precursors of DMS (dimethylsulfoniopropionate) (D6-DMSP) and dimethylsulfoxide ( $^{13}\text{C}$ -DMSO). Results show that de novo biological production of DMS can also take place within brackish melt ponds through bacterial DMSP uptake and cleavage. Our data suggest that FYI melt ponds could represent a reservoir of DMS ranging from ca. 6 to 11 tons of sulfur in the Arctic during July-August available for potential flux to the atmosphere. The importance of this ice-related source of DMS for the Arctic atmosphere is expected to increase as a response to the thinning of sea ice and the areal and temporal expansion of melt ponds on Arctic FYI.

20

25

## 1 Introduction

Melt ponds represent an important but understudied component of the Arctic sea ice system. Snow deposited at the surface of the sea ice progressively melts during the thawing season and may accumulate **above sea level** in depressions at the surface of the ice to form melt ponds (Lüthje et al., 2006), likely through a recently identified process of percolation blockage (Polashenski et al., 2017). In the Arctic, melt pond fraction over first-year sea ice (FYI) in late spring-summer usually ranges from 50 to 60%, locally reaching 90% (Fetterer and Untersteiner, 1998; Eicken et al., 2004; Lüthje et al., 2006; Perovich et al., 2011). Rösel et al. (2012) have reported a 15% increase of the relative melt pond fraction for the month of June during the last decade (2001-2011) in the Arctic, most likely attributable to global climate change. This partly reflects the progressive replacement of multi-year sea ice (MYI) by FYI observed since the 1980's (National Snow and Ice Data Center, NSIDC, <http://nsidc.org>), favouring the formation of shallow melt ponds that spread over increasingly large areas (Agarwal et al., 2011; Ehn et al., 2011). The importance of melt ponds in the Arctic, as a water-air interface involved in heat and gas exchanges, is thus expected to increase in the future.

~~Dimethylsulfide (DMS) is a climate-relevant compound potentially involved in a feedback loop known as the "CLAW" hypothesis (Charlson et al., 1987) linking the biology and climate through the production of DMS-derived sulfate aerosols. Acting as cloud condensation nuclei, the aerosols could contribute to the genesis of longer-lived and clouds with higher albedo (Twomey, 1974; Albrecht, 1989), and thus influence the radiation balance of the Earth. The effect of DMS on cloud properties could be particularly important in remote pristine marine areas such as the polar regions (Carslaw et al., 2013). The summertime Arctic atmosphere displays low coagulation and condensation sinks related to increased wet deposition of particles, and reduced atmospheric aerosol particle loading from anthropogenic sources at lower latitudes (Browse et al., 2012; Croft et al., 2016). Shallow inversion layers that stabilize air masses above the Arctic also reduce the loss of newly formed fine particles. Thermally stable conditions in the marine boundary layer are typical of the Arctic summertime (Tjernström et al., 2012 ; Aliabadi et al., 2016) and associated with clean air masses that allow ultrafine (5–20 nm diameter) particle formation (Burkart et al., 2016), and the potential growth of secondary marine organic aerosols (including DMS-derived particles) into cloud condensation nuclei (Willis et al., 2016). Hence, the Arctic is a favourable terrain for new particle formation from biogenic DMS (Chang et al., 2011; Rempillo et al., 2011 ; Collins et al., 2017; Giamarelou et al., 2016; Mungall et al., 2016; Willis et al., 2016).~~

Dimethylsulfide (DMS) is the main natural source of reduced sulfur for the atmosphere (Bates et al., 1992). Between 17.6 to 34.4 Tg of sulfur are released annually from the ocean to the atmosphere (Lana et al., 2011), accounting for 50-60% of the natural reduced sulfur emitted (Stefels et al., 2007). DMS is also a climate-relevant gas potentially involved in a feedback loop known as the "CLAW" hypothesis (Charlson et al., 1987) linking biology and climate through the production of DMS-derived sulfate aerosols. According to CLAW, DMS emissions may affect the global radiation budget directly through the scattering of incoming solar radiation, and indirectly via the production of cloud condensation nuclei (CCN) leading to the genesis of longer-lived clouds with higher albedo (Twomey, 1974; Albrecht, 1989). Inspiring three decades of

research and hundreds of publications, the feedback mechanism proposed by Charlson et al. (1984) remains yet to be demonstrated in its entirety (e.g. Ayers and Cainey, 2008). Although modelling results show that DMS emissions may have a negative radiative effect (e.g. Bopp et al., 2004; Gunson et al., 2006; Thomas et al., 2010), CCN may exhibit a low sensitivity to changes in DMS on a global scale (Woodhouse et al., 2010). Recent studies questioning the relative importance of DMS in new particle formation have emerged, suggesting that the global CLAW feedback may be weak (e.g. Quinn and Bates, 2011; Green and Hatton, 2014). On a regional scale however, the response of CCN production to change in DMS may vary by a factor of 20 (Woodhouse et al., 2010). The impact of DMS emissions on cloud properties (through the production of CCN) could be particularly important in remote pristine marine areas such as the polar regions (Carslaw et al., 2013). In the Southern Ocean, DMS may have contributed up to 33% of the increase in CCN observed south of 65°S as a response of increased wind speed since the early 1980s (Korhonen et al., 2010). The summertime Arctic marine boundary layer (MBL) is left relatively clean after seasonal wet deposition of particles and reduced atmospheric transport of aerosols from anthropogenic sources at lower latitudes (Stohl, 2006; Browse et al., 2012; Croft et al., 2016). Such pristine conditions, combined with thermally stable MBL are typical of the Arctic summertime (e.g. Aliabadi et al., 2016). Clean Arctic air masses allow ultrafine (5 - 20 nm diameter) particle formation (Burkart et al., 2016), and the potential growth of secondary marine organic aerosols (including DMS-derived particles) into CCN (Willis et al., 2016). Hence, the Arctic is a favourable terrain for new particle formation from biogenic DMS (Chang et al., 2011; Rempillo et al., 2011; Collins et al., 2017; Giamarelou et al., 2016; Mungall et al., 2016; Willis et al., 2016).

DMS produced in the surface oceans represents 95% of the natural reduced sulfur emitted to the atmosphere (Stefels et al., 2007). Between 1 and 40% of the DMSP produced by algae reaches the atmosphere as DMS (Simó and Pedrós-Alió, 1999a). The DMSP-lyase enzymes that mediate DMS production are present in members of the groups Haptophyceae and Dinophyceae, and to a lesser extent, Chrysophyceae (Niki et al., 2000). DMSP plays several roles in phytoplankton, including osmoregulation (Lyon et al., 2016), cryoprotection (Karsten et al., 1996), and prevention of cellular oxidation (Sunda et al., 2002). Two regimes of ocean DMS production are documented. A “bloom-driven” regime in eutrophic regions where the DMS concentrations are controlled by phytoplankton blooms (Stefels et al., 2007), and a “stress-driven” regime in oligotrophic open ocean regions, where DMS concentrations are highly correlated to UV radiation (Toole and Siegel, 2004), nutrient limitation (Stefels, 2000), in situ temperatures (Karsten et al., 1996; Van Rijssel and Gieskes, 2002) and in situ salinity (e.g. Kirst, 1996). Bacteria are also major contributors to DMS production. Part of the DMSP produced by algae is released in the water where it is readily used by heterotrophic bacteria as carbon and sulfur sources (Kiene et al., 2000; Simó, 2001; Vila-Costa et al., 2006). The fraction of dissolved DMSP (DMSP<sub>d</sub>) consumed by heterotrophic bacteria and cleaved into DMS (DMS yield) may vary depending on the microbial community composition, its sulfur requirements, and the availability of other reduced forms of sulfur (Kiene et al., 2000; Stefels et al., 2007). In addition to this pathway, a few studies have demonstrated the potential for reduction of dimethylsulfoxide (DMSO) by marine bacteria and phytoplankton as a source of DMS (e.g. Spiess et al., 2009; Asher et al., 2011). This metabolic pathway is however not ubiquitous among

bacterial assemblages and may not be important quantitatively (Hatton et al., 2012; Hughes et al., 2014). Microbial consumption, photolysis and ventilation are the three sinks influencing the concentrations of DMS in surface mixed layers (Bates et al., 1994; Kieber et al., 1996; Simó and Pedrós-Alió, 1999b; del Valle et al., 2007, 2009).

5 DMS stems mainly from the enzymatic cleavage of dimethylsulfoniopropionate (DMSP) by algal and bacterial DMSP-lyases. DMSP is a cellular metabolite found in several phytoplankton species as particulate DMSP (DMSP<sub>p</sub>) (see the review of Green and Hatton, 2014). DMSP<sub>p</sub> plays various roles in phytoplankton, including osmoregulation (Lyon et al., 2016), cryoprotection (Karsten et al., 1996), and prevention of cellular oxidation (Sunda et al., 2002). Part of the DMSP<sub>p</sub> produced by algae is released in the water column as dissolved DMSP (DMSP<sub>d</sub>) via several pathways reviewed in Stefels et al. (2007), including active exudation, cell lysis, viral lysis and zooplankton grazing. DMSP<sub>d</sub> is then readily available to  
10 heterotrophic bacteria as carbon and sulfur sources (Kiene et al., 2000; Simo, 2001; Vila-Costa et al., 2006). The fraction of DMSP<sub>d</sub> consumed by heterotrophic bacteria and enzymatically cleaved by DMSP-lyases into DMS (DMS yield) may vary depending on the composition of microbial communities, their sulfur requirements, and the availability of other reduced forms of sulfur (Kiene et al., 2000; Stefels et al., 2007). DMSP-lyase enzymes are also present in several members of the microalgal groups Haptophyceae and Dinophyceae, and to a lesser extent Chrysophyceae (Niki et al., 2000). Ultimately,  
15 between ~1 and 40% of the DMSP produced by algae reaches the atmosphere as DMS (Stefels et al., 2007; Simo and Pedros-Alio, 1999a). In addition to the DMSP enzymatic cleavage pathway, DMS production may arise from dimethylsulfoxide (DMSO) reduction by various groups of marine bacteria including proteobacteria (e.g. Vogt et al., 1997), members of the Roseobacter group (Gonzalez et al., 1999) and mat-forming cyanobacteria (van Bergeijk and Stall., 1996). However, the ubiquity of this DMSO-to-DMS reduction pathway amongst bacterial assemblages has not been established  
20 (Hatton et al., 2012). A limited number of phytoplankton species could also be involved in the reduction of DMSO into DMS (e.g. Fuse et al., 1995; Spiese et al., 2009). Increasing evidence suggests that particulate DMSO (DMSO<sub>p</sub>) may be directly synthesized by a potentially wide range of marine phytoplankton (e.g. Lee and de Mora, 1996) and could be involved in osmoprotection, cryoprotection (Lee and de Mora 1999), and anti-oxidant protective mechanisms (Sunda et al., 2002). As for dissolved DMSO (DMSO<sub>d</sub>), it is ubiquitous in seawater and continuous improvements in analytical techniques suggest that  
25 DMSO<sub>d</sub> may be as abundant as DMS in surface waters (e.g. Simo et al., 2000). DMSO is also a known sink for DMS (Hatton et al., 2004) via bacterial and photo oxidation of DMS to DMSO. Vertical Mixing and ventilation are also major removal processes influencing DMS concentrations in surface mixed layers (Bates et al., 1994; Kieber et al., 1996; Simó and Pedrós-Alió, 1999b; del Valle et al. 2007, 2009).

30 Ice-associated environments such as bottom sea ice, brine channels, melt ponds, under-ice surface waters, and leads provide complex and dynamic habitats to diverse microorganism communities involved in sulfur cycling (Levasseur, 2013). In the Arctic, the highest microalgal biomasses are found in the bottom ~0.1 m of sea ice, with Chlorophyll *a* (Chl *a*) concentrations several orders of magnitude above values for under-ice waters values (e.g. Legendre et al. 1992). A similar pattern of DMSP, DMSO and DMS build-up in bottom ice has been reported both in the Arctic and Antarctica (Kirst et al.,

1991; Levasseur et al. 1994; Turner et al., 1995; DiTullio et al., 1995; Lee et al., 2001; Trevena et al., 2003; Trevena and Jones 2006; Delille et al., 2007; Tison et al., 2010; Asher et al., 2011; Nomura et al., 2012; Galindo et al., 2015). For example, DMSP<sub>p</sub> concentrations up to 15 000 nmol l<sup>-1</sup> have been documented during spring in bottom FYI of the Eastern Arctic (Galindo et al., 2014). DMSP, DMSO and DMS are also present throughout the ice column within the brine network (Levasseur et al., 1994; Trevena and Jones, 2006; Asher et al., 2011). Given that primary producers are the sole source of DMSP, very high ice concentrations of Chl *a* are often correlated with DMSP through a first order relationship (Levasseur, 2013). Beyond inter-specific differences in DMSP cellular contents (e.g. Keller et al., 1989; Stefels et al. 2004), environmental forcings are known to control DMSP, DMSO and DMS concentrations. In ice-associated environments, brine volume fraction might also be key in explaining DMS cycling variability via the control of ice permeability (Carnat et al., 2014). The melting season is a pivotal and productive period for these sulfur-containing compounds. Structural changes within sea ice during the melt season, namely increases in brine volume fraction and ice desalination, result in increased connectivity and permeability in the warming sea ice (Willis et al., 2006; Polashenski et al., 2012) and influence DMSP and DMS cycling (Carnat et al., 2014). Also, phytoplankton blooms developing under the ice during the melting period have been shown to produce large quantities of DMSP<sub>p</sub>, potentially leading to a build-up of DMS concentrations (Levasseur et al., 1994). In spite of the spatial importance of melt ponds, only few studies have investigated their role as a source of DMS for the Arctic atmosphere (e.g. Levasseur, 2013; Nomura et al., 2012).

Only three studies have specifically reported on DMS in melt ponds so far. They revealed negligible DMS concentrations in multiyear ice melt ponds in the Central Arctic Ocean, and concentrations ranging from 0.1 to 2.2 nmol l<sup>-1</sup> in the High Arctic (Sharma et al., 1999). In Antarctic melt ponds, DMS concentrations as high as 250 nmol l<sup>-1</sup> were reported by Asher et al. (2011). In the latter study, bacterial DMSO reduction was identified as the main mechanism responsible for the high DMS concentrations. Although the key mechanisms responsible for the DMS measured in Arctic melt ponds has not been assessed so far, their reported colonization by micro-, nano- and pico-sized algae as well as bacteria (Bursa, 1963; Gradinger et al., 2005; Elliott et al., 2015) suggests that DMS in these melt ponds may also originate from algal and bacterial DMSP metabolism. In this study, we report on the DMS concentrations in nine melt ponds located in the Eastern Canadian Arctic Archipelago (CAA), and on the prerequisites and processes responsible for the presence of this climate-active gas.

Considerable efforts have been dedicated to the understanding of underlying process controlling the physics of melt ponds and their feedbacks on climate through the control of surface energy balance of the ice (Lüthje et al., 2006; Polashenski et al., 2017). However, little is known about their biogeochemistry. Four studies have specifically reported on DMS in melt ponds so far. They reveal negligible DMS concentrations in MYI ice melt ponds in the Central Arctic Ocean, and concentrations up to 2.2 nmol l<sup>-1</sup> in the High Arctic (Sharma et al., 1999). In Antarctica, DMS concentrations ranging between 1.1 and 3.7 nmol l<sup>-1</sup> and between below the detection limit (d.l.) and 250 nmol l<sup>-1</sup> were measured in two studies (Nomura et al., 2012 and Asher et al. 2011, respectively). In the latter study, bacterial DMSO reduction was suggested as a possible mechanism responsible for the high DMS concentrations observed although no actual rates of DMS production,

5 either from DMSO or DMSP, were measured. High DMS concentrations reported in the Antarctic are most likely related to the development of a surface ice community following flooding, a process whereby heavy snow load pushes the ice below the water level. Flooding is common in the Antarctic and results in the formation of snow ice (Hunke et al., 2011). Several studies document melt pond colonization by micro-, nano- and pico-sized algae as well as bacteria (Bursa, 1963; Gradinger et al., 2005; Elliott et al., 2015), suggesting that DMS in melt ponds may originate from algal and bacterial metabolism. Yet, in situ DMS production had never been measured nor had key mechanisms been identified. Here, we report on the DMS concentrations in nine melt ponds located in the Eastern Canadian Arctic Archipelago (CAA), and on the prerequisites and processes responsible for the presence of this climate-active gas. This is the first attempt to assess the dynamics of DMS in Arctic melt ponds. We identified sea ice permeability as a major control of DMS production in melt ponds, mediating the transport of both DMS and DMS-producing communities toward the surface of sea ice. We also provide the first evidence for direct in situ DMS production in Arctic melt ponds. We propose that seasonally melting sea ice might become increasingly prone to DMS production as FYI become largely predominant at the regional scale.

## 2 Materials and Methods

### 15 2.1 Study sites and environmental measurements

Nine melt ponds distributed between four stations located in Navy Board Inlet (Ice1 - MP1 and MP2 – 18 July), Barrow Strait (Ice2 - MP1 to MP3 – 20 July, and Ice3 - MP1 and MP2 – 21 July), and Resolute Passage (Ice4 - MP1 and MP2 – 23 July) were sampled during the joint NETCARE/ArcticNet research cruise conducted in 2014 on board the Canadian Coast Guard Ship (CCGS) *Amundsen* (Fig. 1).

20 At each station except for Ice2 (logistical constraints associated with ship time line), measurements of sea ice thickness, snow depth and sea ice freeboard (the height of sea ice above the ocean surface), were conducted within a 3 m distance of the melt ponds using a gauge (Kovacs Enterprise, Roseburg, OR, USA) (Table 1). The 3 m distance was a compromise between maximizing the proximity of ice and melt pond samples and minimizing melt pond disturbance during sampling operations. Ice and freeboard thickness presented in table 1 are averaged values of the 7 (Ice1) to 8 (Ice3 and Ice4) ice cores sampled at each station between the team members for their respective projects. In order to estimate the permeability of the ponded ice, sea ice temperature and bulk salinity were measured following Miller et al. (2015) at stations Ice1, Ice3 and Ice4. Two ice cores for sea ice temperature and salinity measurements were extracted using a 0.09 m core barrel (Kovacs Mark II, Kovacs Enterprise, Roseburg, OR, USA). In situ sea ice temperature profiles were measured directly, at 0.1 m intervals, using a high-precision thermometer (Testo® 720; precision of  $\pm 0.1^\circ\text{C}$ ). Corresponding sea ice salinity profiles were also determined at 0.1 m intervals. Each 0.1 m section was cut with a handsaw, stored in a plastic container, and allowed to melt at room temperature. Bulk salinity of the melted ice section was determined using a conductivity probe (Cond 330i, WTW™; precision of  $\pm 0.1\%$ ). Permeability to fluid transport was assessed with brine

volume profile calculations from bulk salinities and sea ice temperatures following equations from Leppäranta and Manninen (1988) for sea ice temperatures  $> -2^{\circ}\text{C}$  (Fig. 3). Due to logistical constraints mentioned above, neither ice nor snow measurements were conducted at station Ice2.

Melt pond depth, length and width were determined using a graduated stick and a tape ruler. Melt pond water temperature was measured using a high precision thermometer (61220-601 digital data logger, VWR) and water salinity was measured using the conductivity probe mentioned above (Table 2). For each sampling location, two to three members of the research team visually assessed the maximum pond fraction based on pictures taken from the bridge (see Fig. 1c for examples) and a mean value was calculated.

## 2.2 Phytoplankton biomass and enumeration, bacterial count

For Chl *a* quantification, 1000 ml to 1500 ml duplicates of in situ pond water were filtered onto Whatman® GF/F 25 mm filters. Pigments were extracted in 90% acetone for 18 to 24 h in the dark at  $4^{\circ}\text{C}$  (Parsons et al., 1984). Fluorescence of the extracted pigments was measured on board with a Turner Designs fluorometer (model 10-005R; Turner Designs, Inc.) before and after acidification with 5% HCl. The fluorometer was calibrated with a commercially available Chl *a* standard (*Anacystis nidulans*, Sigma). Chl *a* concentrations were calculated using the equation provided by Holm-Hansen et al. (1965).

Microscopic identification and enumeration of eukaryotic cells  $> 2\mu\text{m}$  were conducted in each melt pond. Samples of 250 ml were collected and preserved with acidic Lugol solution (0.4% final concentration; Parsons et al., 1984), then stored in the dark at  $4^{\circ}\text{C}$  until analysis was conducted by inverted microscopy (Lund et al., 1958, Parsons et al., 1984). For each sample, a minimum of 400 cells (accuracy  $\pm 10\%$ ) and three transects of 20 mm were counted at a magnification of 400x. The main taxonomic references used to identify the eukaryotic cells are Tomas and Hasle (1997), Bérard-Therriault et al., (1999) and Thronsdén et al. (2003).

The abundance of bacteria was determined by flow cytometry (Marie et al., 2005). Duplicate 4 ml subsamples were fixed with 20  $\mu\text{l}$  of 25% glutaraldehyde Grade I (0.1% final concentration; Sigma-Aldrich G5882), then subjected to quick-freeze in liquid nitrogen for 24h, and finally stored at  $-80^{\circ}\text{C}$  until analysis. Samples were analyzed using a FACS Calibur FCB3 flow cytometer (Becton Dickinson). Heterotrophic bacteria samples were stained with SYBR Green I and measured at 525 nm to quantify bacteria with Low Nucleic Acid (LNA; potentially less active) and High Nucleic Acid (HNA; potentially more active) content (Gasol and del Giorgio 2000, Lebaron et al. 2001). Analysis were performed on an Epics Altra flow cytometer (Beckman Coulter), fitted with a 488 nm laser (15 mW output; blue), using Expo32 v1.2b software (Beckman Coulter).



### 2.3 DMS(P) sampling, conservation and analysis

Duplicate samples for total DMSP ( $\text{DMSP}_t$ ), dissolved DMSP ( $\text{DMSP}_d$ ) and DMS measurements were collected from the melt ponds using a submersible pump (Cyclone – Aquameric™) connected to a sealed Lead-Acid battery and fitted with LDPE tubing. The pump was placed close to the pond bottom, without touching the ice. Stratified open melt ponds (i.e. melt ponds that have melted all the way to the sea surface) were reported in Arctic FYI (Jung et al., 2015). However, closed FYI melt pond, such as those sampled during this study, are not prone to vertical stratification due to convective- and wind-driven- mixing (Skylvingstad and Paulson, 2007). Given their shallow depths (less than 0.3 m), melt pond stratification was most probably inexistent or minimal during our study. ~~to fill the~~ Glass serum bottles were filled with sampled water, temporarily sealed with a butyl cap and an aluminum lid, and kept in the dark in a cooler until analysis upon return to the ship. Analyses were performed using a purge and trap (PnT) system coupled to a Varian™ 3800 gas chromatograph (GC), equipped with a Pulsed Flame Photometric Detector (PFPD). Analytical precision of the method was better than 5%. Analytical detection limit (d.l.) was  $0.01 \text{ nmol l}^{-1}$  for all sulfur compounds. The protocol is a modified version of the method of Leck and Bågander (1988) as described in Scarratt et al. (2000) and further revised in Lizotte et al. (2012). Briefly, DMS was stripped from liquid samples using helium gas (Praxair™ He, purity 99.999%) flowing at  $50 \pm 5 \text{ ml min}^{-1}$  in the PnT system. One to 5 ml of sample was injected in the PnT. Five ml of MilliQ™ water (Millipore filter system, Millipore Co., Bedford, MA, USA) were subsequently pushed into the system to completely flush the sample into the glass bubbling chamber. The outer walls of the bubbling chamber were heated at  $70^\circ\text{C}$  with a circulating bath. Humidity in the gas sample downstream of the bubbling step was minimized using a  $4^\circ\text{C}$  circulating bath to trigger condensation. A Nafion® membrane separated the gas sample and He-carrier gas from a drying He counter-flow set at  $100 \text{ ml min}^{-1}$  to further desiccate the gas sample. Fluxes in the PnT system were monitored using a flowmeter (Varian™).

For  $\text{DMSP}_t$  samples, 3.5 ml of melt pond water was collected in duplicate into a 5 ml Falcon™ tube. ~~while~~  $\text{DMSP}_d$  was quantified using the less disruptive Small-Volume gravity Drip Filtration (SVDF) method (Kiene and Slezak, 2006). Particulate DMSP ( $\text{DMSP}_p$ ) concentrations were calculated by subtracting  $\text{DMSP}_d$  from  $\text{DMSP}_t$ . DMSP samples were preserved with  $50 \mu\text{l}$  of 50% sulfuric acid ( $\text{H}_2\text{SO}_4$ ) to prevent DMSP transformation and remove pre-existing DMS. Samples were analyzed using the same methods as described above for DMS samples, following mole-to-mole conversion of DMSP into DMS via NaOH (5 M) hydrolysis (Dacey and Blough, 1987).

### 2.4 Process studies

In order to examine the pathways of in situ DMS production in melt ponds, three 24h incubation experiments were conducted with water from the MP1 sampled at stations Ice1, Ice3, and Ice4. Water from the melt ponds was collected using the pump described in sect. 2.3, pooled in clean 19 litres Coleman™ cooler jugs on site, and then transferred into gas-tight 3 litres polyvinyl fluoride Tedlar® bags. Light transmittance through the incubation bag material diminished with decreasing



light wavelength. Between 99 to 92% of the photosynthetically active radiations (PAR, 400-700 nm) were transmitted through the bag material. Transmittances of Ultraviolet A radiations (UVA, 315-400 nm), and Ultraviolet B radiations (UVB, 290-315 nm) ranged between 92 to 82%, and 82 to 38%, respectively. The incubation bags were rinsed once with ~10% HCl, three times with MilliQ™ water, and twice with melt pond water to avoid contamination. The bags were custom-  
5 built and pre-closed on three sides (Dalian Delin Gas Packaging Co., Ltd.). After the addition of the melt pond water, the bags were sealed with Clip-n-seal™ Teflon closure devices. A valve was fitted to each bag to allow the removal of any remaining bubbles.

The samples were subjected to three duplicated treatments (total of 6 bags): 1) two bags of unaltered melt pond water incubated under natural light (Control), 2) two bags amended with D6-DMSP and <sup>13</sup>C-DMSO (100 nmol l<sup>-1</sup>, final  
10 concentration each) incubated under natural light (Light-DMSP/O or L-DMSP/O), and 3) two bags amended with D6-DMSP and <sup>13</sup>C-DMSO (100 nmol l<sup>-1</sup>, final concentration each) incubated in the dark (Dark-DMSP/O or D-DMSP/O). L- and D-DMSP/O bags were amended with ~100 µl of freshly thawed aliquots of two D6-DMSP and <sup>13</sup>C-DMSO stock solutions (high purity >99%, Sigma-Aldrich®). The high concentrations of isotopes added aimed to trigger a rapid biological response (i.e. potential DMS production rates) measurable during our 24h incubations. DMSP and DMSO uptake are not expected to  
15 be mutually exclusive and have been observed concomitantly both in live cultures (Spiese et al., 2009) and in situ (Asher et al., 2011).

Bags were incubated on the foredeck of the ship. The temperature was kept as near to in situ water temperature as possible by continuously flowing surface seawater in the incubator. The temperatures of the incubation water for Ice1-MP1, Ice3-MP1 and Ice4-MP1 were  $1.29 \pm 1.75^\circ\text{C}$ ,  $-0.28 \pm 0.26^\circ\text{C}$ , and  $-0.73 \pm 0.09^\circ\text{C}$ , respectively. These mean values were  
20 within 1°C of the in situ melt pond water temperatures (Table 2).

DMSP, DMSP<sub>d</sub> and DMS concentrations were measured in duplicate every 6h during the incubation period as described above. DMS production from DMSP cleavage and DMSO reduction were determined through GC/mass spectrometry (MS) analysis as an increase of D6-DMS and <sup>13</sup>C-DMS, respectively, in the L-DMSP/O and D-DMSP/O  
25 Treatments. Discrimination by the microorganisms toward lighter (natural) isotopes of DMSP and DMSO is expected to be minimal (< 10%) according to Asher et al. (unpublished data). The observed rates of change in the concentration of DMS stable isotopes are thus assumed to be representative of the potential for DMS cycling in these melt ponds.

This experimental setup allows the measurement of the following rates over 6h and 24h : 1) net changes of in situ DMSP<sub>d</sub> and DMSP<sub>p</sub> in natural light derived from the difference of DMSP<sub>d</sub> and DMSP<sub>p</sub> concentrations versus time in the Controls, respectively, 2) net in situ microbial DMS production in natural light derived from the regression slope of DMS  
30 versus time in the Controls, 3) net potential DMSP<sub>d</sub> changes in natural light and in the dark derived from the regression slope of DMSP<sub>d</sub> versus time in L-DMSP/O and D-DMSP/O, 4) net potential DMS production rate in natural light and in the dark derived from the regression slope of DMS versus time in L-DMSP/O and D-DMSP/O. The daily rates were obtained from

the slopes between final and initial concentrations over 24h. Our experimental setup also allows the estimation of the relative contribution of DMSP and DMSO to the production of DMS, using the discrimination of the different isotopes of DMS (see sect. 2.5).

## 5 2.5 DMS isotopic signatures

The discrimination of the different isotopic forms of DMS, including D6-DMS and  $^{13}\text{C}$ -DMS stemming from D6-DMSP cleavage and  $^{13}\text{C}$ -DMSO reduction, respectively, was performed using GC-MS analysis following purging as described hereafter. Two sets of DMS sample duplicates were taken for the incubation experiments. The first set of duplicates was measured directly on-board using the Varian<sup>TM</sup> 3800 GC described in sect. 2.2. The second set of DMS duplicates was preserved through cryo-trapping. Cryo-trapping of DMS was conducted using glass GC liners filled with Tenax-TA polymer (high sulfur affinity) (Pio et al., 1996; Zemmeling et al., 2002; Pandey and Kim, 2009) kept at  $-80^\circ\text{C}$  prior to their use, and maintained below  $-10^\circ\text{C}$  during the 5 minute purging and trapping process. The Tenax-filled deactivated liners were mounted downstream of the PnT system described earlier. After gas extraction from the liquid samples, Tenax liners and their DMS content were wrapped individually in aluminum foil, placed in a Pyrex<sup>TM</sup> glass tube sealed with a Teflon lid, and returned to the  $-80^\circ\text{C}$  freezer for several weeks until analysis on a land-based GC-MS.

Quantification of D6-DMS and  $^{13}\text{C}$ -DMS was conducted via GC-MS analysis (6978 GC coupled to a 7000B Triple-Quad MS from Agilent). Mass spectra were collected both in full scan ( $m/z$  45–100) and in selected ion monitoring ( $m/z$  62, 63 and 68) modes. Final concentrations were calculated from standard curves using known concentrations of both unlabelled DMS and labelled DMS carrying the D6-DMS and  $^{13}\text{C}$ -DMS signatures. The comparison between fresh DMS samples measured directly on-board during the NETCARE/ArcticNet campaign and cryo-preserved DMS samples shows excellent agreement between the two methods ( $r^2 = 0.96$ , Fig. 2).

## 2.6 Satellite data

Distances between stations Ice1 to Ice4 and the open ocean were assessed using scaled NASA's Earth Observing System Data and Information System (EOSDIS) imagery. Maps of the ice cover were accessed for the sampling dates in July 2014 through the MODIS (Terra/Aqua) Corrected Reflectance (True Color) layer combined with MODIS (Terra) Corrected Reflectance (Bands 3,6,7). These data are accessible in open source through the Global Imagery Browse Services (GIBS) (<https://worldview.earthdata.nasa.gov>). The imagery had a resolution of 250 m on a daily scale.

## 30 2.7 Statistical Analysis

Normality of the data was assessed using the Shapiro-Wilk test with a 0.05 significance level (R statistical software, R Core Team, 2016), which revealed that most variables were non-normally distributed ( $n=9$ ,  $df=8$ ,  $\alpha=0.05$ ). Non-parametric Spearman's rank correlation test ( $r_s$ ) with a 0.05 significance level was used to assess correlation between key variables since normality could not be achieved uniformly through standard normalization methods ( $n=9$ ,  $df=8$ ,  $\alpha=0.05$ ). Model I linear regressions ( $r^2$ ) were used to determine biological rates during the incubation experiments (Sokal and Rohlf, 1995).

A non-parametric Mann-Whitney U test was used to determine whether the distributions of reduced-sulfur compounds (i.e. DMS,  $DMSP_p$  and  $DMSP_d$ ) in the Ice1-MP1 and Ice4-MP1 incubations experiments were statistically different from one another. The difference in reduced sulfur compound concentrations between the two incubation experiments was not found to be statistically significant ( $n=45$ ,  $df=16$   $\alpha=0.05$ ).

Based on the results of the Mann-Whitney U test, a series of Wilcoxon Signed-rank tests with a significance level  $\alpha=0.05$  were conducted on the combined datasets in order to 1) assess the presence of statistical differences between the Controls and each Treatment L-DMSP/O and D-DMSP/O; 2) assess the potential effect of light on the concentrations and change rates of the reduced sulfur compounds under study (DMS,  $DMSP_d$  and  $DMSP_p$ ) by comparing paired dependent samples (repeated measures) from L-DMSP/O and D-DMSP/O.

## 3 Results

### 3.1 Ponded sea ice and snow properties

The physical characteristics of the sea ice surrounding the melt ponds are presented in table 1 and in figure 3. All the sampling sites were characterized by FYI, which was the predominant ice type throughout the region under study. Averaged sea ice thickness around the melt ponds were relatively uniform, varying between  $1.13 \pm 0.07$  and  $1.27 \pm 0.01$  m at the different sites. Average freeboard values were relatively more variable. Station Ice1 was characterized by low ice freeboards  $-0.01 \pm 0.01$  m. Station Ice3 had the highest positive freeboards with  $0.10 \pm 0.02$  m. Station Ice4 freeboards were also positive but showed the greatest variability, with  $0.07 \pm 0.04$  m.

In order to estimate the permeability of the ponded sea ice, in situ temperature and bulk salinity of the full-depth sea ice were measured within 5 m distance of the melt ponds. Based on the "rule of five's", these temperature and salinity values suggest that the top sea ice was permeable around the melt ponds (Golden et al., 1998). Brine volume fraction was calculated using sea ice salinity and temperature values, and used as a proxy of sea ice permeability (Fig. 3). Averaged values for bulk sea ice salinity over the full thickness of the ice were 1.73, 2.83 and 3.75 at stations Ice1, Ice3 and Ice4, respectively. Maximum bulk salinity never exceeded 5.00 (Ice4, 1.2-1.3 m section). In situ temperatures, averaged over the full thickness of the ice, were  $-0.54$  °C,  $-0.52$  °C and  $-0.98$  °C at stations Ice1, Ice3 and Ice4, respectively, and reached a minimum value of  $-1.39$  °C (Ice4, 0.8-0.9 m section). Brine volume fraction constantly exceeded 10% in the ice profiles, except in the upper 0.1 m section of the Ice3 station where we likely observed the effects of refreezing metamorphosis of snow and/or sea ice

recrystallization. Snow meltwater percolation and refreezing can form superimposed fresh ice layers as observed at station Ice3. The resulting impermeable layer at the top of the ice contributed to the high freeboard (0.1 m) measured at this station, representing 6% of the total ice thickness. Visual estimates of maximum pond fraction ranged from 30 to 60% (see Fig. 1c) and the remaining surface of sea ice was bare ice at stations Ice1, Ice2 and Ice4.

### 5 3.2 Physical, chemical and biological characteristics of the melt pond water

The physical and chemical characteristics of the melt ponds are presented in table 2. All melt ponds were closed melt ponds, i.e. not directly connected with the water column (Lee et al., 2012). The mean depth of the individual melt ponds ranged from 0.07 to 0.29 m, with length and width varying between 1.00 and 25.00 m (Fig. 1). Melt pond water temperatures and salinities varied between 0.21 and 1.86°C and between 0.2 and 8.5, respectively. Chl *a* concentrations were variable, ranging from 0.03 to 0.48 µg l<sup>-1</sup> with a mean of 0.20 µg l<sup>-1</sup> (Table 3). The composition of the algal assemblage present in the melt ponds will be described in details in a companion paper but is summarized in table 3. The algal assemblages were dominated by unidentified flagellates, ice-associated pennate diatoms, and chrysophytes. Empty diatom frustules were abundant in all melt ponds. Abundance of heterotrophic bacteria with high nucleic acid content (HNA) varied between 0.02 and 0.24 × 10<sup>9</sup> cells l<sup>-1</sup> (Table 3).

15 In situ DMSP<sub>p</sub> and DMSP<sub>d</sub> concentrations ranged from 1.8 to 4.0 nmol l<sup>-1</sup>, and from below d.l. to 1.4 nmol l<sup>-1</sup>, respectively. Melt pond DMS concentrations ranged from below detection limit (d.l., < 0.01 nmol l<sup>-1</sup>) to 6.1 nmol l<sup>-1</sup> (Table 3). Spearman's rank correlation coefficients between key in situ variables measured in the melt ponds are presented in table 4. DMS concentrations significantly co-varied with salinity ( $r_s = 0.84$ ,  $p < 0.05$ ) and Chl *a* ( $r_s = 0.84$ ,  $p < 0.05$ ). None of the other variables measured displayed significant relationships between each other.

20

### 3.3 Dynamics/cycling of reduced sulfur compounds in Arctic melt ponds

Results from the Ice1-MP1 and Ice4-MP1 incubation experiments are presented in Fig. 4 (4a-c left and 4b-d right, respectively). Results from the Ice3-MP1 experiments are not shown since DMSP<sub>d</sub> and DMS concentrations showed no variation during the 24h incubation period in the Controls and in the Amended Treatments. This will be discussed in sect.

25 4.2.2.

During the Ice1-MP1 incubation, initial DMSP<sub>d</sub> concentration was 1.30 nmol l<sup>-1</sup> in the Control and slightly increased to reach 5.3 nmol l<sup>-1</sup> during the 24h incubation period (Fig. 4a). In the Light (L-DMSP/O) and Dark (D-DMSP/O) Amended Treatments, DMSP<sub>d</sub> concentrations started at 102 nmol l<sup>-1</sup>, decreased to ~ 35 nmol l<sup>-1</sup> at T<sub>6</sub>, and remained stable (Dark Treatments) or decreased to 10 nmol l<sup>-1</sup> (Light Treatments) until T<sub>24</sub> (Fig. 4a). Concentrations of DMS in the Control of Ice1-MP1 started at 3.0 nmol l<sup>-1</sup>, increased to 8.8 nmol l<sup>-1</sup> between T<sub>0</sub> and T<sub>6</sub>, and then decreased regularly to 4.2 nmol l<sup>-1</sup> at T<sub>24</sub> (Fig. 4c). The addition of labelled DMSP and DMSO stimulated DMS production. In the L-DMSP/O Treatment, DMS

30

concentrations increased to  $12.6 \text{ nmol l}^{-1}$  at  $T_6$ , remained at this level between  $T_6$  and  $T_{12}$ , increased again between  $T_{12}$  and  $T_{18}$  and remained stable at  $\sim 19 \text{ nmol l}^{-1}$  between  $T_{18}$  and  $T_{24}$  (Fig. 4c). DMS concentrations were consistently higher in the D-DMSP/O Treatment than in L-DMSP/O (Fig. 4c). They first reached  $15.6 \text{ nmol l}^{-1}$  at  $T_6$ , increased gradually to reach a peak value of  $24.2 \text{ nmol l}^{-1}$  at  $T_{18}$ , and decreased slightly to  $21.6 \text{ nmol l}^{-1}$  at  $T_{24}$ . Note that dissolved DMSO was not measured during this study due to methodological issues.

In the Ice4-MP1 incubation,  $\text{DMSP}_d$  concentrations started at  $3.0 \text{ nmol l}^{-1}$  in the Control and remained close to this value during the whole experiment (Fig. 4b). In the L-DMSP/O and D-DMSP/O Amended Treatments,  $\text{DMSP}_d$  concentrations started at  $87$  and  $96 \text{ nmol l}^{-1}$ , respectively. As observed in the previous melt pond, the concentrations decreased to  $\sim 45 \text{ nmol l}^{-1}$  at  $T_6$ , and then slowly decreased to a value of  $\sim 30 \text{ nmol l}^{-1}$  at  $T_{24}$  (Fig. 4b). DMS concentrations in the Control of Ice1-MP1 started at  $2.6 \text{ nmol l}^{-1}$  and remained at this level during the 24h experiment (Fig. 4d). In the L-DMSP/O Treatment, DMS concentrations increased more or less linearly from  $2.6 \text{ nmol l}^{-1}$  at  $T_0$  to  $6.7 \text{ nmol l}^{-1}$  at  $T_{24}$ . In the D-DMSP/O Treatment, the increase in DMS concentrations was steeper than in the Light Treatment, and a maximal value of  $11.5 \text{ nmol l}^{-1}$  was reached at  $T_{24}$ .

In situ and potential change rates of the sulfur compounds during the incubation experiments are presented in tables 5 and 6, respectively. Changes in  $\text{DMSP}_d$  and to a lesser extent DMS concentrations were generally not linear over the 24h incubation period, with more pronounced variations during the first 6 h. To take into account this non-linearity, both hourly rates measured between  $T_0 - T_6$  and  $T_6 - T_{24}$ , as well as daily rates ( $T_0 - T_{24}$ ) are presented in these tables.

In Ice1-MP1, the concentrations of  $\text{DMSP}_p$  in the Control decreased at a rate of  $2.2 \text{ nmol l}^{-1} \text{ d}^{-1}$  (Table 5). We measured no change in  $\text{DMSP}_d$  during the first 6 h, but a positive net increase of  $4.0 \text{ nmol l}^{-1}$  over the full 24h incubation period was observed. In situ DMS changes show an increase rate of  $1.0 \text{ nmol l}^{-1} \text{ h}^{-1}$  during the first 6 h and of  $1.2 \text{ nmol l}^{-1} \text{ d}^{-1}$  over 24 h. Potential net  $\text{DMSP}_d$  change rates of  $-11.6$  and  $-10.2 \text{ nmol l}^{-1} \text{ h}^{-1}$  were measured during the first 6 h of incubation in L- and D-DMSP/O Treatments, respectively (Table 6). These rates became  $-1.2$  and  $-0.6 \text{ nmol l}^{-1} \text{ h}^{-1}$  between  $T_6$  and  $T_{24}$  in L- and D-DMSP/O, respectively. Over 24 h, negative potential net  $\text{DMSP}_d$  change rates of  $\sim -91 \text{ nmol l}^{-1}$  and  $-71 \text{ nmol l}^{-1}$  for the L-DMSP/O and D-DMSP/O Treatments were calculated. Positive potential net DMS change rates of  $1.6$  and  $2.1 \text{ nmol l}^{-1} \text{ h}^{-1}$  were measured during the first 6 h of incubation in L-DMSP/O and D-DMSP/O, respectively. For the complete 24h incubation, potential net DMS change rates reached  $15.4 \text{ nmol l}^{-1} \text{ d}^{-1}$  in the Light and  $18.6 \text{ nmol l}^{-1} \text{ d}^{-1}$  in the Dark.

In Ice4-MP1, in situ  $\text{DMSP}_p$  decreased at a rate of  $1.9 \text{ nmol l}^{-1} \text{ d}^{-1}$  over the course of the incubation (Table 5). Meanwhile, in situ  $\text{DMSP}_d$  changes rates were below the d.l. during the first 6 h and almost null over 24 h (Table 5). In situ DMS change rates were close to zero after 6 h, and below d.l. after 24 h. Potential net  $\text{DMSP}_d$  change rates of  $-8.1 \text{ nmol l}^{-1} \text{ h}^{-1}$  were measured during the first 6 h of incubation in both L- and D-DMSP/O (Table 6). These rates slowed down to  $-0.5$  and  $-0.9 \text{ nmol l}^{-1} \text{ h}^{-1}$  between  $T_6$  and  $T_{24}$ , respectively. Over one day, average potential net  $\text{DMSP}_d$  change rates of  $\sim -59 \text{ nmol l}^{-1}$

and  $-62 \text{ nmol l}^{-1}$  were calculated for the L-DMSP/O and D-DMSP/O Treatments. Potential net DMS change rates remained low in both L-DMSP/O and D-DMSP/O Treatments during the first 6 h of incubation with values at  $0.1$  and  $0.3 \text{ nmol l}^{-1} \text{ h}^{-1}$ , respectively. For the complete 24h incubation, potential net DMS change rates in Light and Dark reached  $4.2$  and  $8.9 \text{ nmol l}^{-1} \text{ d}^{-1}$ , respectively.

5            During both Ice1-MP1 and Ice4-MP1 incubation experiments, the Light versus Dark Treatment had no effect on the net changes in DMSP<sub>d</sub> concentrations between the L-DMSP/O and D-DMSP/O Treatments (Wilcoxon Signed-rank test;  $n=8$ ,  $df=3$ ,  $\alpha=0.05$ ), but significantly impacted the rates of net accumulation of DMS (Wilcoxon Signed-rank test;  $n=12$ ,  $df=5$ ,  $\alpha=0.05$ ). The accumulation of DMS over 24h in the L-DMSP/O Treatments were consistently and significantly lower than in the corresponding D-DMSP/O Treatments (Wilcoxon Signed-rank test;  $n=8$ ,  $df=3$ ,  $\alpha=0.05$ ). Based on the difference between  
10 the L- and D-DMSP/O Treatments after 24 h, we estimated the light-associated DMS sinks at  $3.2 \text{ nmol l}^{-1} \text{ d}^{-1}$  in Ice1-MP1 and at  $4.7 \text{ nmol l}^{-1} \text{ d}^{-1}$  in Ice4-MP1 (Table 6).

### 3.4 Isotopic discrimination of DMS sources

Table 7 shows the concentrations of DMS isotopes ( $m/z$  62) and ( $m/z$  68) after 24h incubation in the three incubation  
15 treatments and their relative contribution (%) to the total DMS measured at  $T_{24}$ . As expected, 100% of the total DMS in the Controls of these two experiments ( $3.0 \text{ nmol l}^{-1}$  and  $2.3 \text{ nmol l}^{-1}$ ) showed the isotopic signature of natural DMS ( $m/z$  62). In the L-DMSP/O Treatment of the Ice1-MP1 incubation, 78% ( $14.4 \text{ nmol l}^{-1}$ ) of the DMS measured at  $T_{24}$  derived from D6-DMSP additions ( $m/z$  68), with the remaining 22% ( $4.1 \text{ nmol l}^{-1}$ ) being natural DMS ( $m/z$  62) (Table 5). Similarly, 73% ( $18.2 \text{ nmol l}^{-1}$ ) of the DMS measured at  $T_{24}$  derived from D6-DMSP additions ( $m/z$  68) in the D-DMSP/O Treatment, with  
20 the remaining 27% ( $6.6 \text{ nmol l}^{-1}$ ) carrying the signature of natural DMS ( $m/z$  62).

In Ice4-MP1, 80% ( $5.1 \text{ nmol l}^{-1}$ ) of the DMS measured at  $T_{24}$  in the L-DMSP/O Treatment derived from the added D6-DMSP, with the remaining 20% ( $1.3 \text{ nmol l}^{-1}$ ) carrying the signature of natural DMS ( $m/z$  62). For the D-DMSP/O Treatment, 65% ( $7.9 \text{ nmol l}^{-1}$ ) of the DMS at  $T_{24}$  derived from the D6-DMSP addition ( $m/z$  68) with 35% ( $4.2 \text{ nmol l}^{-1}$ ) originating from natural DMS. The absence of ( $m/z$  63) DMS, regardless of the treatment, indicates that  $^{13}\text{C}$ -DMSO  
25 reduction was not contributing to the production of DMS during these two experiments ( $m/z$  63 not shown in table 7). The match between the sum of DMS isotopes ( $m/z$  62 and  $m/z$  68) and the total fresh DMS concentration measured on board (Fig. 2) also confirms the absence of DMSO-to-DMS reduction during our experiments.

## 4 Discussion

Research on DMS dynamics in melt ponds is in its infancy. Before this study, only four publications reported DMS  
30 measurements in melt ponds, two in the Arctic (Leck and Persson, 1996; Sharma et al., 1999) and the two others in the Antarctic (Asher et al., 2011; Nomura et al., 2012). In the Arctic, Leck and Persson reported negligible levels of DMS in

MYI melt ponds while Sharma et al. (1999) measured concentrations reaching 2.2 nmol l<sup>-1</sup>. In the Antarctic, Nomura et al. reported DMS concentrations inferior to 3.7 nmol l<sup>-1</sup> while Asher et al. (2011) measured levels up to 250 nmol l<sup>-1</sup>. Our current limited understanding of the mechanisms responsible for the cycling of DMS in melt ponds prevents the identification of the underlying causes of these differences. Our results show that DMS concentrations in Arctic melt ponds may be at least three times higher (up to 6 nmol l<sup>-1</sup>) than the first Arctic measurements and that both physical and biological processes can contribute to the accumulation of this climate-active gas in these transient environments. As discussed hereafter, our results suggest that different ice cover dynamics and microbial communities are the two probable leading causes for the reported variability in DMS concentrations between melt ponds.

#### 4.1 Physical controls of DMS concentrations in melt ponds

The strong relationship observed between DMS concentrations and salinity in the melt ponds sampled ( $r_s = 0.84$ ,  $p \leq 0.05$ , Table 4) suggests that salinization processes may play a crucial role in the initial seeding of DMS (and probably DMS-producing microbial assemblages) and the resulting cycling of DMS within melt ponds. Three main mechanisms could be involved in the salinization of closed melt ponds: 1) deposition of sea spray from the ice margin/leads, 2) ice brine intrusion, and 3) seawater intrusion through porous/low freeboard sea ice. For the reasons explained below, seawater intrusion through porous/low freeboard sea ice appears to be the most likely mechanism responsible for the salinization of the melt ponds during our study.

Sea spray probably did not contribute significantly to the salinization of the melt ponds during our study. The salinization of melt ponds could occur through sea spray deposition or seawater overflow during stormy events. Sea spray can transport salts over distances ranging from a few meters for the largest particles to a maximum distance of ~ 30 km for finer aerosols, depending on wind speed (McArdle and Liss, 1995). This requires favourable wind direction, a relative proximity of the melt ponds with open water areas, and as demonstrated hereafter regarding the melt ponds studied here, unrealistic volumes of sea spray. During our study, the average volume of the melt ponds was 8 m<sup>3</sup>. We conservatively estimated that 19 to 367 litres of sea spray (assuming an average sea surface salinity of 33) was required to increase melt pond salinity from zero to 0.2 or 8.5, as measured during our study. Considering both the relatively large volume of sea spray required and the far-reaching distances (>15 km, estimated from MODIS data) of the sampled melt ponds from open water at the time of sampling, sea spray was unlikely the main source of salt in the melt ponds studied.

Ice brine intrusion is also unlikely to have contributed significantly to melt pond salinization since the averaged bulk ice salinity was low (under 5), especially in the top 0.2 m where it did not exceed 2. It is also known that most of the hyper-saline brine characterizing consolidated cold FYI in winter are lost in spring through full depth brine convection well before melt ponds start to form (Jardon et al., 2013). Residual salts are finally lost through meltwater flushing during the summer season (Weeks and Ackley, 1986, Eicken et al., 2002; Vancoppenolle et al., 2007). At the time of our sampling, low bulk salinity values, combined with calculated brine volume fraction constantly exceeding 10% in the entire sea ice profiles

(except in the upper 0.1 m section of the Ice3 station) suggest that full depth flushing had already occurred. We thus exclude sea ice brine enrichment of melt ponds as their main salinization mechanism.

5 Rather, we suggest that melt ponds salinization originated mostly from the intrusion of seawater through the ice. Although closed melt ponds are not visibly connected to seawater, exchanges with the underlying seawater can take place. The extent of these exchanges are dependent on the sea ice freeboard and micro-structure, i.e. the amount, size and shape of brine inclusions (Carnat et al., 2014), that controls sea ice permeability. Above a critical brine volume ranging between 5% (for columnar sea ice) and 10% (for granular sea ice), brine inclusions become interconnected. During the melting season, decrease in sea ice thickness is enhanced by the formation of the melt pond and lead to a loss of freeboard. As melt ponds become closely levelled with seawater, small changes in ice temperature oscillating around the freezing temperature may result in episodic intrusion of seawater mixed with meltwater through the porous ice. Seawater mixed with meltwater penetrating the brines channels of permeable sea ice may bring salts, nutrients and microorganisms (Jardon et al., 2013, Vancoppenolle et al., 2010), potentially reaching surface melt ponds. This mechanism most probably explains the salinity and biochemical characteristics of Ice1 and Ice4 melt ponds. Station Ice3 represents a different case. Here, the low melt pond salinity (and absence of biological activity) may be explained by the presence of an impermeable ice layer on the top of the ice preventing both pond drainage and exchange between pond water and seawater.

15 We acknowledge that our data set is too limited to draw firm conclusions on the processes governing the formation and salinization of FYI melt ponds. Yet, in the interest of further research, we conjecture that snow load before melt onset may be crucial in determining the fate of melt ponds not only with regards to their saline status, but also their potential to produce DMS. Brine volume, derived from bulk salinity and temperature, generally provides a valid proxy for sea ice permeability. In some case however, melting of high snowpack generates a considerable flow (up to 15cm d<sup>-1</sup>) of freshwater into the porous structure of sea ice (Polashenski et al., 2017). This can create localized ice plugs within the highly connected brine network of apparently porous sea ice and allow melt ponds to persist above sea level well after sea ice bulk sea ice brine volume reached a critical level (5-10%). Such deviation from the porosity/permeability relationship following freshwater intrusion is demonstrated in Polashenski et al. (2017). We suggest that we observed such case of melt pond persistence above sea level in station Ice3. Alternatively, lower snow load remaining at the onset of the melt season will translate into a less abundant freshwater input above sea ice. Snow load distribution is however notoriously highly variable even at the meter scale due to wind redistribution and sea ice topography variability (e.g. Polashenski et al., 2017). Low snowpack would induce limited insulation of the sea ice from atmospheric conditions, resulting in 1) a more gradual warming of sea ice during spring season, and 2) limited freshwater loading available for percolation blockage. In such case, freshwater would not seal the ice through percolation blockage (Polashenski et al., 2017). Sea ice would then remain entirely porous as soon as the 5-10% brine volume threshold is reached, facilitating melt pond salinization process. We suggest that this scenario may have been observed at stations Ice1 and Ice4.



## 4.2 Biological control of DMS production in melt ponds

### 4.2.1 Simulated in situ conditions

In addition to the physical mechanisms mentioned above, results from our incubation experiments show that biological production of DMS may take place in Arctic melt ponds under simulated in situ conditions, and to a higher extent following DMSP enrichment. A significant daily net DMS production of  $1.2 \text{ nmol l}^{-1} \text{ d}^{-1}$  was measured without substrate addition in one of the three melt ponds tested, Ice1-MP1 (Table 5). The absence of net daily increase in DMS in the two other melt ponds tested does not necessarily preclude potential gross production since, as discussed below, this production could be balanced by microbial DMS uptake and photolysis. Such balance between DMS sources and sinks over a 24h period has been previously observed during incubation experiments conducted with Labrador Sea water (Wolfe et al., 1999). However, this explanation probably does not explain the absence of accumulation of DMS in the freshwater melt pond Ice3-MP1 since the addition of substrate failed to stimulate DMS production.

### 4.2.2 Source of DMS under substrate amended conditions

Bacterial DMSP<sub>a</sub> metabolism was the main mechanism underlying DMS production in the melt ponds tested. None of the DMS measured carried the (m/z 63) isotopic signature that would have indicated its <sup>13</sup>C-DMSO origin. The absence of DMSO reduction during our incubation contrasts with the results of Asher et al. (2011) who concluded that this process was the main pathway driving extremely high gross DMS production rates within sea ice brines (up to  $105 \pm 24 \text{ nmol l}^{-1} \text{ d}^{-1}$ ), as well as high concentrations of DMS ( $250 \text{ nmol l}^{-1}$ ) in melt ponds in Antarctica. Extremely high gross DMS production rates from DMSO reduction, up to  $105 \pm 24 \text{ nmol l}^{-1} \text{ d}^{-1}$ , were measured within Antarctic sea ice brines by Asher et al. (2011). The authors suggested that this mechanism could also potentially be responsible for the high DMS concentrations (up to  $250 \text{ nmol l}^{-1}$ ) measured in Antarctic melt ponds. The absence of DMS production from <sup>13</sup>C-DMSO in the melt ponds in our study may then reflect potential differences in microbial assemblages within melt ponds, as the metabolic ability to convert DMSO into DMS is not ubiquitous among bacterial communities (Hatton et al., 2012; Hughes et al., 2014). In support of this hypothesis, it has been shown that between 70 and 78% of the operational taxonomic units (OTU), a marker of microbial diversity, in Arctic and Southern Ocean surface water communities are unique to their region (Ghiglione et al., 2012). Observed differences in the biological characteristics of melt ponds between the poles could also reflect divergent sea ice dynamics. Antarctic sea ice salinity is higher by 0.5 to 1.0% than in Arctic sea ice (Gow et al., 1982, 1987) and the C-shaped salinity profile that is typical in fully formed Arctic FYI is not as prominent in Southern Ocean sea ice (Eicken, 1992). Antarctic sea ice is commonly subjected to intense rafting. Flooding, a process whereby heavy snow load pushes the ice below the water level, is common in the Antarctic and results in the formation of snow ice (Hunke et al., 2011). Antarctic melt ponds studied in Asher et al. (2011) may have been subjected to this flooding leading to the formation of salted “freeboard layers” (Haas et al., 2001; Massom et al., 2006). This is supported by the reported highest salinities in the top sea

ice layers and the subsequent salinity decrease throughout the ice profile (Asher et al. 2011). Such configuration may bring highly productive microbial communities at the surface of the ice, potentially responsible for the high DMS concentrations observed in melt ponds. The still limited availability of data, including other published studies, prevents us from firmly concluding on the specific reasons of the absence of DMS production from  $^{13}\text{C}$ -DMSO and compels additional exploration.

5

#### 4.2.3 Substrate limitation of microbial DMSP uptake and DMS production

The addition of DMSP had a strong stimulating effect on the bacterial uptake of DMSP and the resulting production of DMS in the two brackish melt ponds tested. In both Ice1-MP1 and Ice4-MP1, the response of the microbial assemblage to the addition of DMSP was rapid and strong (Fig. 4) as approximately half of the  $\text{DMSP}_d$  added was consumed over the first 6 h and potential net DMS production increased substantially.

10

In the Amended Treatments, changes in the  $\text{DMSP}_d$  concentrations over time proceeded into two distinct phases during the incubation period (Figs. 4a-c). Irrespective of the light regime, the first phase ( $T_0$  to  $T_6$ ) was characterized by a rapid net decrease of  $\text{DMSP}_d$  concentrations. Potential net  $\text{DMSP}_d$  change rates of  $\sim -11 \text{ nmol l}^{-1} \text{ h}^{-1}$  in Ice1-MP1 and of  $-8.1 \text{ nmol l}^{-1} \text{ h}^{-1}$  in Ice4-MP1 (Table 6) were calculated. These estimates represent minimum rates since our calculation assumes a linear uptake during the first 6 h. Even so, these rates already translate an extremely steep decrease of  $\text{DMSP}_d$  in comparison with those of  $-0.01$  to  $-0.2 \text{ nmol l}^{-1} \text{ h}^{-1}$  previously measured in the same region in the water column and under the ice cover in spring (Luce et al., 2011; Galindo et al., 2015). This difference most probably reflects the large amount of DMSP added in our experiments. The second phase of the incubation (from  $T_6$  to  $T_{24}$ ) shows an abrupt slowing down of the potential net  $\text{DMSP}_d$  change rates, still slightly superior but closer to the range of in situ rates reported by the previous studies (Table 6). These results clearly show that an active microbial assemblage predisposed to  $\text{DMSP}_d$  consumption inhabited the brackish melt ponds under study. This is in accordance with Sørensen et al. (2017) reported substrate limitation of bacterial growth in Arctic FYI melt ponds.

15

20

The bi-phasic DMSP uptake dynamics observed in our experiment suggests that DMSP additions at least temporarily fulfilled the microbial requirement for this substrate. Phytoplankton biomass, and probably dissolved organic carbon, was low in the melt ponds. In this context of substrate limitation, rapid uptake of  $\text{DMSP}_d$  was expected. Fast and transient intracellular accumulation of compatible solutes, such as DMSP, may serve as an adaptive strategy by microbial cells to help cope with fluctuations of the surrounding environment, increasing their tolerance to osmotic and thermal stresses for example (Welsh, 2000). Such accumulations which could occur under replete conditions allow a so-called “luxury uptake” of compounds by microorganisms above their immediate requirements. Finally, the low HNA bacterial abundances measured in the melt ponds (Table 3) might explain the curtailing of  $\text{DMSP}_d$  uptake measured after the initial rapid consumption.

25

30

Following DMSP<sub>d</sub> addition, the potential daily net DMS production rates varied between 4.2 and 18.6 nmol l<sup>-1</sup> in the two brackish melt ponds tested (Table 6). As previously mentioned, it was only within the freshwater Ice3-MP1 melt pond that potential to process DMSP and produce DMS was not detected, even when substrate limitation was alleviated by DMSP<sub>d</sub> addition. These different in situ and potential DMSP metabolisms and DMS production rates suggest that de novo  
5 DMS production in melt ponds is triggered only once a threshold in microbial biomass is reached. In support of this hypothesis, Chl *a* concentration (0.05 µg l<sup>-1</sup>) and bacterial abundance (0.02 x 10<sup>9</sup> cells l<sup>-1</sup>) were extremely low in the unproductive freshwater Ice3-MP1: one order of magnitude lower than in the two productive brackish Ice1-MP1 and Ice4-MP1 (Table 3).

In contrast with the simulated in situ conditions in the Controls, net potential DMS production in the Amended  
10 Treatments constantly exceeded DMS loss through photolysis and bacterial consumption, resulting in a net accumulation of DMS throughout the 24 h of incubation (Fig. 4c, d). In spite of the atypically high DMSP level added, our DMSP<sub>d</sub> amendments could be considered as analogues of the DMSP<sub>d</sub> pulses that take place in the natural environment during the senescence phase of algal blooms, or under high viral attack and grazing pressure. These pulses are known to contribute to transient DMS build-up at lower latitudes (e.g. Malin et al., 1993; Locarnini et al., 1998; Scarratt et al., 2000). At high  
15 latitudes, the inhibitory effect of low temperature on microbial DMS consumption may even exacerbate these build-ups. For instance, temperatures below 2°C were found to potentially inhibit DMS consumption rates in the Labrador Sea (Wolfe et al., 1999). The sensitivity of DMS microbial uptake to low temperature was proposed by Wolfe et al. (1999) as a potential driving mechanism responsible for the large pulses of DMS often measured in the Arctic environment. Cold and biologically active melt ponds may thus be prone to such DMS accumulation when the limitation in substrate is alleviated. However, our  
20 observations suggest that such events, that would require high biomass, may be rare in Arctic melt ponds.

#### 4.4.4 Influence of light on DMSP bacterial metabolism

Light affected the accumulation of DMS in the DMSP/O Amended Treatments. The continuous light conditions prevailing during our incubation experiments reduced DMS accumulation in the L-DMSP/O Treatments compared to the D-DMSP/O  
25 Treatments by ~15% and up to 40% in Ice1-MP1 and Ice4-MP1, respectively (Fig. 4c, d). This negative effect of light was expected since photolysis is known as an important sink for DMS in the open ocean, sometimes as important as bacterial consumption in the near surface waters (Royer et al., 2016). However, removing light did not increase DMSP<sub>d</sub> removal rates (Fig. 4a, b). It should be pointed out that our incubation setup did not aim to reproduce the exact light field of the melt ponds where light backscattering could considerably increase DMS loss by photolysis. The importance of light as a sink for DMS  
30 in melt ponds should be thoroughly investigated in future studies. Light-induced DMS losses may be particularly relevant in melt ponds since DMS ventilation, another important sink for DMS (absent from our incubation setup), is probably limited at least in small melt ponds where fetch is minimal.

## 5 Conclusion

Results from this study confirm the presence of DMS in Arctic melt ponds, with concentrations up to three times higher than those reported by the two other previous Arctic studies. Salinization of melt ponds appears to be a prerequisite to the presence of DMS and its de novo biological production. Intrusion of seawater through porous sea ice and low freeboard flooding seems to be a fundamental mechanism for bringing salt and DMS in the melt ponds as well as allowing the establishment of potential DMS-producing communities. As melt ponds become closely levelled with seawater, small changes in ice temperature oscillating around the freezing temperature may result in episodic intrusion of seawater mixed with meltwater through the porous ice. Seawater mixed with meltwater penetrating the brines channels of permeable sea ice may bring salts, nutrients and microorganisms potentially seeding surface melt ponds. Results from our incubation experiments reveal a modest but measurable in situ net production of DMS in one of the melt ponds tested. Our data also suggests that melt ponds can host an active bacterial assemblage associated with rapid DMSP uptake when available and significant daily production of DMS. Freshwater ponds lacked the potential to produce DMS, further confirming the importance of the seawater intrusion mechanism in the biological cycling of DMS in melt ponds. No DMSO-to-DMS reduction was detected in our study. A

To this day, most climatologies assume the absence of DMS fluxes above ice-covered waters (e.g. Lana et al., 2011) even though several studies provide direct (Zemmelink et al., 2008; Nomura et al., 2012, MYI) and indirect (Carnat et al., 2014, FYI) evidence of DMS venting from snow-covered Antarctic sea ice. Arctic studies have also reported DMS exchanges above the ice-covered ocean, specifically highlighting the importance of particular zones such as open leads (Levasseur et al., 1994) and cracks in sea ice, as well as melt ponds (Sharma et al., 1999; Mungall et al., 2016). Here, we measured an average DMS concentration of 2.1 nmol l<sup>-1</sup> in nine FYI melt pond. Although estimation of the actual DMS flux from the melt ponds sampled here is beyond the scope of our study, we argue that FYI melt ponds represent a significant reservoir of DMS in the Arctic readily available for air-sea exchange. The estimation of the importance of melt ponds as net sources of DMS for the atmosphere will require an accurate evaluation of their spatial and temporal coverage, a better understanding of gas exchange between small fetch melt ponds and the atmosphere and its sensitivity to changing wind velocity, as well as comprehensive measurements of DMS within melt ponds at large, both FYI and MYI, and particularly at higher latitudes. How the strength of DMS emissions from melt ponds will respond to changes in Arctic climate is still unknown. Both the spatial extent of melt ponds and their temporal span have increased over the last three decades in connection with regional climate alterations (Stroeve et al., 2014; Agarwal et al., 2011). Meanwhile, MYI is increasingly being replaced by thinner FYI (e.g. Kwok et al., 2009), potentially promoting melt pond salinization processes through permeable sea ice. The importance of this ice-related source of DMS for the Arctic atmosphere could increase as a response of these structural changes of the Arctic ecosystem.

## Data availability

Metadata are available on the Polar Data Catalog website at [www.polardata.ca](http://www.polardata.ca). Data are available on request by contacting the first author.

## Authors contribution

- 5 Margaux Gourdal was responsible for the elaboration of the experimental design, the sampling process, the data analysis and processing, and the redaction of this paper. Several co-authors provided specific data included in the paper and all co-authors contributed to the final edition of the paper.

## Competing interests

The authors declare that they have no conflict of interest.

## 10 Special issue statement

We are requesting your permission to link this paper to a NETARE BG special issue.

## Acknowledgements

- The authors wish to thank the commandant, officers, and crew of the Canadian ice-breaker NGCC *Amundsen* for their support during the project. The authors are especially indebted to Jean-Sébastien Côté, Tim Papakyriakou and Roghayeh  
15 Ghahremaninezhad for participating to the sample collection, Marjolaine Blais for pigment and bacterial abundance analysis, and Virginie Galindo for her extensive logistical support during the cruise. We thank Sylvie Lessard for cell identification. This project was funded by the Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments (NETCARE), ArcticNet (Network of Centres of Excellence of Canada), the Canada Excellence Research Chair in Remote Sensing of Canada's New Arctic Frontier, and the Takuvik Joint International Laboratory. Partial funding  
20 was also provided by the Natural Sciences and Engineering Research Council of Canada (NSERC) and the Fonds de Recherche du Québec Nature et Technologies (FRQNT) through Québec-Océan. Funding support was also received from the Canadian Museum of Nature for cell counts analysis. The author received graduate scholarships from Université Laval, [takuvik UMI](#) and stipends from NETCARE and Québec-Océan.

## References

- 25 Agarwal, S., Moon, W., and Wettlaufer, J. S.: Decadal to seasonal variability of Arctic sea ice albedo, *Geophys. Res. Lett.*, 38, L20504, doi:10.1029/2011GL049109, 2011.
- [Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, \*Science\*, 245\(4923\), 1227-1231, 1989.](#)

- Aliabadi, A. A., Staebler, R. M., de Grandpré, J., Zadra, A., and Vaillancourt, P. A.: Comparison of Estimated Atmospheric Boundary Layer Mixing Height in the Arctic and Southern Great Plains under Statically Stable Conditions: Experimental and Numerical Aspects, *Atmos.-Ocean*, 54, 60–74, doi:10.1080/07055900.2015.1119100, 2016.
- 5 Asher, E. C., Dacey, J. W., Mills, M. M., Arrigo, K. R., and Tortell, P. D.: High concentrations and turnover rates of DMS, DMSP and DMSO in Antarctic sea ice, *Geophys. Res. Lett.*, 38, L23609, doi: 10.1029/2011GL049712, 2011.
- Ayers, G. P., and Cainey, J. M.: The CLAW hypothesis: a review of the major developments, *Environmental Chemistry*, 4(6), 366-374, 2008.
- Bates, T. S., Kiene, R. P., Wolfe, G. V., Matrai, P. A., Chavez, F. P., Buck, K. R., Blomquist, B., W., and Cuhel, R. L.: The cycling of sulfur in surface seawater of the northeast Pacific, *J. of Geophys. Res.-Oceans*, 99(C4), 7835-7843, 1994.
- 10 Bérard-Therriault, L., Poulin, M, and Bossé, L.: Guide d'identification du phytoplancton marin de l'estuaire et du golfe du Saint-Laurent, incluant également certains protozoaires, *Publ. spéc. can. sci. halieut. Aquat*, 128, 1-387, 1999.
- Bopp, L., Boucher, O., Aumont, O., Belviso, S., Dufresne, J. L., Pham, M., and Monfray, P.: Will marine dimethylsulfide emissions amplify or alleviate global warming? A model study, *Canadian Journal of Fisheries and Aquatic Sciences*, 61(5), 826-835, 2004.
- 15 Bouillon, R. C., Lee, P. A., de Mora, S. J., Levasseur, M., and Lovejoy, C.: Vernal distribution of dimethylsulphide, dimethylsulphonioacetate, and dimethylsulphoxide in the North Water in 1998. *Deep Sea Research Part II: Topical Studies in Oceanography*, 49(22-23), 5171-5189, 2002.
- Browse, J., Carslaw, K. S., Arnold, S. R., Pringle, K., and Boucher, O.: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, *Atmos. Chem. Phys.*, 12, 6775–6798, doi:10.5194/acp-12-6775-2012, 2012.
- 20 Burkart, J., Willis, M. D., Bozem, H., Thomas, J. L., Law, K., Hoor, P., et al.: Summertime observations of elevated levels of ultrafine particles in the high Arctic marine boundary layer, *Atmospheric Chemistry and Physics*, 17(8), 5515, 2017.
- Bursa, A.: Phytoplankton in coastal waters of the Arctic Ocean at Point Barrow, Alaska. *Arctic*, 16(4), 239-262, 1963.
- 25 Carnat, G. et al.: Investigations on physical and textural properties of Arctic first-year sea ice in the Amundsen Gulf, Canada, November 2007 – June 2008 (IPY-CFL system study), *Journal of Glaciology*, 59(217), 819-837, 2013.
- Carnat, G. et al.: Physical and biogeochemical controls on DMSP dynamics in ice shelf influenced fast ice during a winter-spring and a spring-summer transitions, *Journal of Geophysical Research-Oceans*, 119(5), 2882-2905 2014.
- Carslaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., Mann, G.W., Spracklen, D.V, Woodhouse, M.T., Regayre, L.A. and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature*, 503(7474), 67-71, 2013.
- 30

- Chang, R. Y. W., Sjostedt, S. J., Pierce, J. R., Papakyriakou, T. N., Scarratt, M. G., Michaud, S., Levasseur, M., Leaitch, W. R. and Abbatt, J. P. D.: Relating atmospheric and oceanic DMS levels to particle nucleation events in the Canadian Arctic, *J. Geophys. Res.-Atmos.* 116(D00S03), doi, 10.1029/2011JD015926, 2011.
- Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G.: Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, *Nature*, 326(6114), 655-661, 1987.
- Collins, D. B., Burkart, J., Chang, R. Y. W., Lizotte, M., Boivin-Rioux, A., Blais, M., Mungall, E., L., Boyer, M., Irish, V., E., Massé, G., Kunkel, D., Tremblay, J-É, Papakyriakou, T., Bertram, A., K., Bozem, H., Gosselin, M., Levasseur, M., Abbatt, J., P., D.: Frequent Ultrafine Particle Formation and Growth in the Canadian Arctic Marine Environment, *Atmos. Chem. Phys. Discuss.*, doi.org/10.5194/acp-2017-411, 2017.
- 10 Cox, G. F. and Weeks, W. F.: Equations for determining the gas and brine volumes in sea ice samples, *Journal of Glaciology*, 29(102), 306-316, 1983.
- Croft, B., Martin, R. V., Leaitch, W. R., Tunved, P., Breider, T. J., D'Andrea, S. D. and Pierce, J. R.: Processes controlling the annual cycle of Arctic aerosol number and size distributions, *Atmos. Chem. Phys.*, 16(6), 3665–3682, doi:10.5194/acp-16-3665-2016, 2016.
- 15 Dacey, J. W. and Blough, N. V.: Hydroxide decomposition of dimethylsulfoniopropionate to form dimethylsulfide, *Geophysical Research Letters*, 14(12), 1246-1249, 1987.
- Delille, B., Jourdain, B., Borges, A. V., Tison, J. L., and Delille, D.: Biogas (CO<sub>2</sub>, O<sub>2</sub>, dimethylsulfide) dynamics in spring Antarctic fast ice, *Limnology and Oceanography*, 52(4), 1367-1379, 2007.
- del Valle, D. A., Kieber, D. J., and Kiene, R. P.: Depth-dependent fate of biologically-consumed dimethylsulfide in the Sargasso Sea, *Mar. Chem.*, 103(1), 197-208, 2007.
- 20 del Valle, D. A. D., Kieber, D. J., Toole, D. A., Brinkley, J., and Kiene, R. P.: Biological consumption of dimethylsulfide (DMS) and its importance in DMS dynamics in the Ross Sea, Antarctica, *Limnol. and Oceanogr.*, 54(3), 785-798, 2009.
- De Mora, S. J. et al.: Aspects of the biogeochemistry of sulphur in glacial melt water ponds on the McMurdo Ice Shelf, Antarctica, *Antarctic Science*, 8(1), 15-22, 1996.
- 25 Derksen, C., Piwowar, J. and LeDrew, E.: Sea ice melt-pond fraction as determined from low level aerial photographs, *Arctic and Alpine Research*, 345-351, 1997.
- Dickson, D.M.J., Wyn Jones, R.G., and Davenport, J.: Steady state osmotic adaptation in *Ulva lactuca*. *Planta*, 150, 158-165, doi: 10.1007/BF00582360, 1980.

- DiTullio, G. R., and Smith Jr, W. O.: Relationship between dimethylsulfide and phytoplankton pigment concentrations in the Ross Sea, Antarctica, Deep Sea Research Part I: Oceanographic Research Papers, 42(6), 873-892, 1995.
- Drinkwater, M. R., and Lytle, V. I.: ERS 1 radar and field-observed characteristics of autumn freeze-up in the Weddell Sea, J. Geophys. Res.-Oceans., 102(C6), 12593-12608, doi:10.1029/97JC00437, 1997.
- 5 Ehn, J. K., Mundy, C. J., Barber, D. G., Hop, H., Rossnagel, A. and Stewart, J.: Impact of horizontal spreading on light propagation in melt pond covered seasonal sea ice in the Canadian Arctic, J. Geophys. Res., 116, C00G02, doi:10.1029/2010JC006908, 2011.
- Eicken, H.: Salinity profiles of Antarctic sea ice: Field data and model results, J. Geophys. Res., 97, 15,545–15,557, 1992.
- 10 Eicken, H., Grenfell, T. C., Perovich, D. K., Richter-Menge, J. A., and Frey, K.: Hydraulic controls of summer Arctic pack ice albedo, J. Geophys. Res.-Oceans., 109, C08007, doi:10.1029/2003JC001989, 2004.
- Eicken, H., Krouse, H. R., Kadko, D. and Perovich, D. K., Tracer studies of pathways and rates of meltwater transport through arctic summer sea ice, J. Geophys. Res., 107(C10), 8046, doi:10.1029/2000JC000583, 2002.
- Elliott, A., Mundy, C. J., Gosselin, M., Poulin, M., Campbell, K., and Wang, F.: Spring production of mycosporine-like amino acids and other UV-absorbing compounds in sea ice-associated algae communities in the Canadian Arctic, 15 Mar. Ecol. Prog. Ser., 541, 91-104, doi:10.3354/meps11540, 2015.
- Fetterer, F. and Untersteiner, N.: Observations of melt ponds on Arctic sea ice, J. Geophys. Res.-Oceans., 103(C11), 24821-24835, doi: 10.1029/98JC02034, 1998.
- Flocco, D., Schröder, D., Feltham, D. L. and Hunke, E. C.: Impact of melt ponds on Arctic sea ice simulations from 1990 to 2007, J. Geophys. Res.117, C09032, 2012.
- 20 Fuse, H., Takimura, O., Kamimura, K., Murakami, K., Yamaoka, Y., and Murooka, Y.: Transformation of dimethyl sulfide and related compounds by cultures and cell extracts of marine phytoplankton, Bioscience, biotechnology, and biochemistry, 59(9), 1773-1775, 1995.
- Galindo, V., M. Levasseur, C. J. Mundy, M. Gosselin, J.-E. Tremblay, M. Scarratt, Y. Gratton, T. Papakiriakou, M. Poulin, and M. Lizotte, Biological and physical processes influencing sea ice, under-ice algae, and 25 dimethylsulfoniopropionate during spring in the Canadian Arctic Archipelago, J. Geophys. Res. Oceans, 119, 3746–3766, doi:10.1002/2013JC009497, 2014.
- Galindo, V., Levasseur, M., Scarratt, M., Mundy, C. J., Gosselin, M., Kiene, R. P., Gourdal, M. and Lizotte, M.: Under-ice microbial dimethylsulfoniopropionate metabolism during the melt period in the Canadian Arctic Archipelago, Mar. Ecol. Prog. Ser., 524, 39-53, doi:10.3354/meps11144, 2015.



- Gasol, J. M., and Del Giorgio, P. A.: Using flow cytometry for counting natural planktonic bacteria and understanding the structure of planktonic bacterial communities, *Sci. Mar.*, 64(2), 197-224,2000.
- 5 Ghiglione, J. F., Galand, P. E., Pommier, T., Pedrós-Alió, C., Maas, E. W., Bakker, K., .et al.: Pole-to-pole biogeography of surface and deep marine bacterial communities, *Proceedings of the National Academy of Sciences*, 109(43), 17633-17638, 2012.
- Giamarelou, M., Eleftheriadis, K., Nyeki, S., Tunved, P., Torseth, K., and Biskos, G.: Indirect evidence of the composition of nucleation mode atmospheric particles in the high Arctic, *J. of Geophys. Res -Atmos.*, 121(2), 965-975, 2016.
- Golden, K. M.: Critical behavior of transport in sea ice, *Physica B*, 338 (1), 274–283, 2003.
- Golden, K. M., Ackley, S. F., and Lytle, V. I.: The percolation phase transition in sea ice, *Science*, 282(5397), 2238-2241. doi:10.1126/science.282.5397.2238, 1998.
- 10 González, J. M., Kiene, R. P., and Moran, M. A.: Transformation of Sulfur Compounds by an Abundant Lineage of Marine Bacteria in the  $\alpha$ -Subclass of the Class Proteobacteria, *Applied and environmental microbiology*, 65(9), 3810-3819, 1999.
- Gow, A. J., Ackley, S. F., Buck, K. R., and Golden, K. M.: Physical and structural characteristics of Weddell Sea pack ice, Tech. Rep. 87-14, CRREL Cold Reg. Res. Eng. Lab., Hanover, N.H, 1987.
- 15 Gow, A. J., Ackley, S. F., Weeks, W. F., and Govoni, J. W.: Physical and structural characteristics of Antarctic sea ice, *Ann. Glaciol.*, 3, 113–117, 1982.
- Gradinger, R. R., Meiners, K., Plumley, G., Zhang, Q., and Bluhm, B. A.: Abundance and composition of the sea-ice meiofauna in off-shore pack ice of the Beaufort Gyre in summer 2002 and 2003, *Polar Biol.*, 28(3), 171-181, doi: 10.1007/s00300-004-0674-5, 2005.
- 20 Granger, J., D., M., Sigman, M., D., Prokopenko, Lehmann, M., F., and Tortell, P. D: A method for nitrite removal in nitrate N and O isotope Analysis, *Limnol. Oceanogr. Methods* 4, 205–212, 2006.
- Green, T. K. And Hatton, A. D.: The CLAW hypothesis: a new perspective on the role of biogenic sulphur in the regulation of global climate, *Oceanography and Marine Biology, An Annual Review*, 52, 315-336, 2014.
- 25 Gunson, J. R., Spall, S. A., Anderson, T. R., Jones, A., Totterdell, I. J., and Woodage, M. J.: Climate sensitivity to ocean dimethylsulphide emissions, *Geophysical Research Letters*, 33(7), 2006.
- Haas, C., Thomas, D. N., and Bareiss, J.: Surface properties and processes of perennial Antarctic sea ice in summer, *J. Glaciol.*, 47(159), 613-625, 2001.
- Hatton AD, Darroch L, Malin G: The role of dimethylsulphoxide in the marine biogeochemical cycle of dimethylsulphide, *Oceanogr Mar Biol Annu Rev*, 42:29–56, 2004.
- 30

- Hatton, A. D., Shenoy, D. M., Hart, M. C., Mogg, A., and Green, D. H.: Metabolism of DMSP, DMS and DMSO by the cultivable bacterial community associated with the DMSP-producing dinoflagellate *Scrippsiella trochoidea*, *Biogeochemistry*, 110(1-3), 131-146, doi: 10.1007/s10533-012-9702-7, 2012.
- 5 Holm-Hansen, O., Lorenzen, C. J., Holmes, R. W., and Strickland, J. D.: Fluorometric determination of chlorophyll ICES, *J. Mar. Sci.*, 30(1), 3-15, 1965.
- Hunke, E. C., Notz, D., Turner, A. K., and Vancoppenolle, M.: The multiphase physics of sea ice: a review for model developers, *The Cryosphere*, 5, 989-1009, <https://doi.org/10.5194/tc-5-989-2011>, 2011.
- Jardon, F. P., Vivier, F., Vancoppenolle, M., Lourenço, A., Bouruet-Aubertot, P., and Cuypers, Y. Full-depth desalination of warm sea ice, *J. Geophys. Res.-Oceans.*, 118(1), 435-447, doi:10.1029/2012JC007962, 2013.
- 10 Jung, J., Min, J. O., Yang, E. J., Lee, S. H., and Kang, S. H.: Nutrients in melt ponds and snows on Arctic sea ice during 2014 sea ice camp, 2015 (Only abstract is available).
- Karsten, U., Kück, K., Vogt, C., and Kirst, G. O. (Eds.): Dimethylsulfoniopropionate production in phototrophic organisms and its physiological functions as a cryoprotectant. In *Biological and environmental chemistry of DMSP and related sulfonium compounds*, Springer, New York, USA, 1996.
- 15 Kattner, G., Thomas, D. N., Haas, C., Kennedy, H., and Dieckmann, G. S.: Surface ice and gap layers in Antarctic sea ice: highly productive habitats, *Mar. Eco. Prog. Series*, 277, 1-12, 2004.
- Kieber, D. J., Jiao, R. P., Kiene, R. P., and T. S. Bates: Impact of dimethylsulfide photochemistry on methyl sulfur cycling in the equatorial Pacific Ocean, *J. Geophys. Res.*, 101(C2), 3715–3722, doi:10.1029/95JC03624, 1996.
- Kiene, R. P., Linn, L. J., and Bruton, J. A., New and important roles for DMSP in marine microbial communities, *J. Sea Res.*, 43(3), 209-224, 2000.
- 20 Kiene, R. P., and Slezak, D.: Low dissolved DMSP concentrations in seawater revealed by small-volume gravity filtration and dialysis sampling, *Limnol. and Oceanogr.: Methods*, 4(4), 80-95, 2006.
- Kirst, G. O., Thiel, C., Wolff, H., Nothnagel, J., Wanzek, M., and Ulmke, R.: Dimethylsulfoniopropionate (DMSP) in icealgae and its possible biological role, *Mar. Chem.*, 35(1-4), 381-388, doi: 10.1016/S0304-4203(09)90030-5, 25 1991.
- Kirst, G. O. (Eds): Osmotic adjustment in phytoplankton and macroalgae. In *Biological and environmental chemistry of DMSP and related sulfonium compounds*, Springer, New York, USA, 1996.
- Korhonen, H., Carslaw, K. S., Forster, P. M., Mikkonen, S., Gordon, N. D. and Kokkola, H.: Aerosol climate feedback due to decadal increases in Southern Hemisphere wind speeds, *Geophys. Res. Lett.*, 37, L02805, 30 [doi:10.1029/2009GL041320](https://doi.org/10.1029/2009GL041320), 2010.

- Kovacs, A.: Sea ice. Part 1. Bulk salinity versus ice floe thickness (No. CRREL-96-7). Cold regions research and engineering lab Hanover NH, 1996.
- Lana, A., T. G. Bell, T. G., Simó, R., Vallina, S. M., Ballabrera-Poy, J., Kettle, A. J., Dachs, J., Bopp, L., Saltzman, E. S., Stefels, J., Johnson, J. E. And Liss, P. S.: An updated climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean, *Global Biogeochem. Cy.*, 25, GB1004, doi:10.1029/2010GB003850, 2011.
- Landy, J., Ehn, J., Shields, M. and Barber D.: Surface and melt pond evolution on landfast first-year sea ice in the Canadian Arctic Archipelago, *J. Geophys. Res. Oceans*, 119, 3054–3075, doi:10.1002/2013JC009617, 2014.
- Lebaron, P., Servais, P., Agogué, H., Courties, C., and Joux, F.: Does the high nucleic acid content of individual bacterial cells allow us to discriminate between active cells and inactive cells in aquatic systems?, *Appl. and Environ. Microbiol.*, 67(4), 1775-1782, 2001.
- Leck, C., and Bågander, L. E.: Determination of reduced sulfur compounds in aqueous solutions using gas chromatography-flame photometric detection, *Anal. Chem.*, 60(17), 1680-1683, doi: 10.1021/ac00168a011, 1988.
- Leck, C. and Persson, C.: The central Arctic Ocean as a source of dimethyl sulfide: seasonal variability in relation to biological activity, *Tellus B*, 48(2), 156-177, 1996.
- Lee, P. A., and De Mora, S. J.: Intracellular dimethylsulfoxide (DMSO) in unicellular marine algae: speculations on its origin and possible biological role, *Journal of Phycology*, 35(1), 8-18, 1999.
- Lee, P. A., De Mora, S. J., Gosselin, M., Levasseur, M., Bouillon, R. C., Nozais, C., and Michel, C.: Particulate dimethylsulfoxide in Arctic sea-ice algal communities: The cryoprotectant hypothesis revisited, *Journal of Phycology*, 37(4), 488-499, 2001.
- Lee, S. H., Stockwell, D. A., Joo, H.-M., Son, Y. B., Kang, C.-K. and Whitledge, T. E.: Phytoplankton production from melting ponds on Arctic sea ice, *J. Geophys. Res.*, 117, C04030, doi:10.1029/2011JC007717, 2012.
- Legendre, L., Ackley, S. F., Dieckmann, G. S., Gulliksen, B., Horner, R., Hoshiai, T., et al.: Ecology of sea ice biota, *Polar Biology*, 12(3-4), 429-444, 1992.
- Leppäranta, M., and Manninen, T.: The brine and gas content of sea ice with attention to low salinities and high temperatures, *Internal Rep.*, pp. 1 – 14, *Finn. Inst. of Mar. Res.*, Helsinki, 1988.
- Levasseur, M.: Impact of Arctic meltdown on the microbial cycling of sulphur, *Nature Geoscience*, 6(9), 691, 2013.
- Levasseur, M., Gosselin, M., and Michaud, S.: A new source of dimethylsulfide (DMS) for the arctic atmosphere: ice diatoms, *Mar. Biol.*, 121(2), 381-387, 1994.
- Levasseur, M., Scarratt, M. G., Michaud, S., Merzouk, A., Wong, C. S., Arychuk, M., Richrdson, W., Rivkin, R. B., Hale., M. Wong, E. Marchetti, A. and Kiyosawa, H.: DMSP and DMS dynamics during a mesoscale iron fertilization

experiment in the Northeast Pacific—Part I: Temporal and vertical distributions, *Deep-Sea Res. Pt. II*, 53(20), 2353-2369, 2006.

- 5 Lizotte, M., Levasseur, M., Michaud, S., Scarratt, M. G., Merzouk, A., Gosselin, M., and Kiene, R. P.: Macroscale patterns of the biological cycling of dimethylsulfoniopropionate (DMSP) and dimethylsulfide (DMS) in the Northwest Atlantic, *Biogeochemistry*, 110(1-3), 183-200, doi:10.1007/s10533-011-9698-4, 2012.
- Locarnini, S. J. P., Turner, S. M., and Liss, P. S.: The distribution of dimethylsulfide, DMS, and dimethylsulfoniopropionate, DMSP, in waters off the Western Coast of Ireland. *Cont. Shelf Res.*, 18(12), 1455-1473, 1998.
- Luce, M., Levasseur, M., Scarratt, M. G., Michaud, S., Royer, S. J., Kiene, R., Lovejoy, C., Gosselin, M., Poulin, M., Gratton, Y. and Lizotte, M.: Distribution and microbial metabolism of dimethylsulfoniopropionate and dimethylsulfide during the 2007 Arctic ice minimum, *J. Geophys. Res.-Oceans.*, 116(C00G06), doi:10.1029/2010JC006914, 2011.
- 10 Lund, J. W. G., Kipling, C., and Le Cren, E. D.: The inverted microscope method of estimating algal numbers and the statistical basis of estimations by counting, *Hydrobiologia*, 11(2), 143-170, 1958.
- Lüthje, M., Feltham, D. L., Taylor, P. D., and Worster, M. G.: Modeling the summertime evolution of sea-ice melt ponds, *J. Geophys. Res.-Oceans.*, 111(C02001), doi:10.1029/2004JC002818, 2006.
- 15 Lyon, B.R. et al.: Role of dimethylsulfoniopropionate as an osmoprotectant following gradual salinity shifts in the sea-ice diatom *Fragilariopsis cylindrus*, *Environmental Chemistry*, 13(2), 181-194, 2016.
- Malin, G., Turner, S., Liss, P., Holligan, P., and Harbour, D.: Dimethylsulphide and dimethylsulphoniopropionate in the Northeast Atlantic during the summer coccolithophore bloom, *Deep-Sea Res. Pt. I*, 40(7), 1487-1508, doi: 10.1016/0967-0637(93)90125-M, 1993.
- 20 Marie D, Simon N, Vaultot D (Eds.), *Phytoplankton cell counting by flow cytometry*. Andersen R., *Algal culturing techniques*, Academic Press, London, U.K., 2005.
- Markus, T., J. C. Stroeve, and J. Miller.: Recent changes in Arctic sea ice melt onset, freezeup, and melt season length, *J. Geophys. Res.*, 114, C12024, doi:10.1029/2009JC005436, 2009.
- 25 Massom, R. A., Eicken, H., Hass, C., Jeffries, M. O., Drinkwater, M. R., Sturm, M., Worby, A. P., Wu, X, Lytle, I., Ushio, S., Morris, K., Reid, P.A., Warren, S. G., and Allison. I.: Snow on Antarctic sea ice, *Rev. Geophys.*, 39(3), 413-445, 2001.
- Massom, R. A., Stammerjohn, S. E., Smith, R. C., Pook, M. J., Iannuzzi, R. A., Adams, N., et al.: Extreme anomalous atmospheric circulation in the West Antarctic Peninsula region in austral spring and summer 2001/02, and its profound impact on sea ice and biota, *Journal of climate*, 19(15), 3544-3571, 2006.
- 30

- McArdle, N. C., and Liss, P. S.: Isotopes and atmospheric sulphur, *Atmos. Environ.*, 29(18), 2553-2556, doi:10.1016/1352-2310(95)00188-5, 1995.
- Miller, L. A. et al.: Methods for biogeochemical studies of sea ice: The state of the art, caveats, and recommendations, *Elem. Sci. Anth.*, 3, 2015.
- 5 Morris, K., and Jeffries, M. O.: Seasonal contrasts in snow-cover characteristics on Ross Sea ice floes, *Ann. Glaciol.*, 33(1), 61-68, 2001.
- Mungall, E. L., Croft, B., Lizotte, M., Thomas, J. L., Murphy, J. G., Levasseur, M., Randall V. Martin R.V., Wentzell, J.J.B., Liggio, J. and Abbatt, J. P.: Dimethyl sulfide in the summertime Arctic atmosphere: measurements and source sensitivity simulations, *Atmos. Chem. Phys.*, 16(11), 6665-6680, 2016.
- 10 Nakawo, M., and Sinha, N. K., Growth rate and salinity profile of first-year sea ice in the high Arctic, *J. Glaciol.*, 27, 315-331, 1981.
- Niki, T., Kunugi, M., and Otsuki, A.: DMSP-lyase activity in five marine phytoplankton species: its potential importance in DMS production, *Mar. Biol.*, 136(5), 759-764, 2000.
- Nomura, D. et al.: Direct measurements of DMS flux from Antarctic fast sea ice to the atmosphere by a chamber technique, *Journal of Geophysical Research Oceans*, 117(C4), 2012.
- 15 Notz, D.: Thermodynamic and fluid-dynamical processes in sea ice, Ph. D. thesis, University of Cambridge, 2005.
- Notz, D. and Worster, M. G.: Desalination processes in sea ice, *J. Geophys. Res.*, 114, C05006, doi: 10.1029/2008JC004885, 2009.
- Pandey, S. K., and Kim, K. H.: A review of methods for the determination of reduced sulfur compounds (RSCs) in air. *Environ. Sci. Technol.* 43(9), 3020-3029, doi:10.1021/es803272f, 2009.
- 20 Parsons, T. R., Maita, Y., and Lalli, C. M.: A manual of biological and chemical methods for seawater analysis, Publ. Pergamon Press, Oxford, 1984.
- Perovich, D. K., Richter-Menge, J. A., Jones, K. F., Light, B., Elder, B. C., Polashenski, C., Laroche, D., Markus, T. and Lindsay, R.: Arctic sea-ice melt in 2008 and the role of solar heating, *Ann. Glaciol.*, 52(57), 355-359, doi:10.3189/172756411795931714, 2011.
- 25 Pio, C. A., Castro, L. M., Cerqueira, M. A., Santos, I. M., Belchior, F., and Salgueiro, M. L.: Source assessment of particulate air pollutants measured at the southwest European coast., *Atmos. Environ.*, 30(19), 3309-3320. doi: 10.1016/1352-2310(96)00058-1, 1996.

- Polashenski, C., Golden, K. M., Perovich, D. K., Skyllingstad, E., Arnsten, A., Stwertka, C., and Wright, N.: Percolation blockage: A process that enables melt pond formation on first year Arctic sea ice, *J. of Geophys. Res.-Oceans*, 122(1), 413-440, 2017.
- 5 Polashenski, C., Perovich, D., and Courville, Z.: The mechanisms of sea ice melt pond formation and evolution, *Journal of Geophysical Research: Oceans*, 117(C1), 2012.
- Quinn, P. K. and Bates, T. S.: The case against climate regulation via oceanic phytoplankton sulphur emissions, *Nature*, 480(7375), 51, 2011.
- R Core Team: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria, <https://www.R-project.org/>, 2016.
- 10 Rempillo, O., Seguin, A. M., Norman, A.-L., Scarratt, M., Michaud, S., Chang, R., Sjostedt, S., Abbatt, J., Else, B., Papakyriakou, T., Sharma, S., Grasby, S. and Levasseur, M.: Dimethyl sulfide air-sea fluxes and biogenic sulfur as a source of new aerosols in the Arctic fall, *J. Geophys. Res.*, 116, D00S04, doi:10.1029/2011JD016336, 2011.
- Rösel, A., Kaleschke, L., and Birnbaum, G.: Melt ponds on Arctic sea ice determined from MODIS satellite data using an artificial neural network, *The Cryosphere*, 6(2), doi:10.5194/tc-6-431-2012, 431, 2012.
- 15 Royer, S. J., Galí, M., Mahajan, A. S., Ross, O. N., Pérez, G. L., Saltzman, E. S., and Simó, R.: A high-resolution time-depth view of dimethylsulphide cycling in the surface sea, *Sci. Rep.-UK.*, 6, 32325, 2016.
- Scarratt, M. G., Levasseur, M., Schultes, S., Michaud, S., Cantin, G., Vezina, A., and De Mora, S. J.: Production and consumption of dimethylsulfide (DMS) in North Atlantic waters, *Mar. Ecol. Prog. Ser.*, 204, 13-26, doi:10.3354/meps204013, 2000.
- 20 Scharien, R. K., Segal, R., Nasonova, S., Nandan, V., Howell, S. E. L., and Haas, C.: Winter Sentinel-1 backscatter as a predictor of spring Arctic sea ice melt pond fraction. *Geophysical Research Letters*, 44, 12,262–12,270. doi:10.1002/2017GL075547, 2017.
- Schröder, D., Feltham, D. L., Flocco, D., and Tsamados, M.: September Arctic sea ice minimum predicted by spring melt-pond fraction, *Nature Climate Change*, 4(5), 353, 2014.
- 25 Sharma, S., Barrie, L.A., Plummer, D., McConnell, J.C., Brickell, P.C., Levasseur, M., Gosselin, M. and Bates, T.S.: Flux estimation of oceanic dimethyl sulfide around North America, *J. Geophys. Res.*, 104, 21327-21342, 1999.
- Sharp, Z.: *Principles of Stable isotope Geochemistry*, Prentice Hall, New York. 360 pp. , 2007.
- Simó, R.: Production of atmospheric sulfur by oceanic plankton: biogeochemical, ecological and evolutionary links, *Trends Ecol. Evol.*, 16(6), 287-294, 2001.

- Simó, R., and Pedrós-Alió, C.: Role of vertical mixing in controlling the oceanic production of dimethyl sulphide, *Nature* 402:396–399, 1999a.
- Simó, R., and Pedrós-Alió, C.: Short-term variability in the open ocean cycle of dimethylsulfide, *Global Biogeochem. Cy.*, 13, 1173–1181, 1999b.
- 5 Simó, R., Pedrós-Alió, C., Malin, G., and Grimalt, J. O.: Biological turnover of DMS, DMSP and DMSO in contrasting open-sea waters, *Mar. Ecol. Prog. Ser.*, 203, 1-11, 2000.
- Skyllingstad, E. D., and Paulson, C. A.: A numerical study of melt ponds, *J. Geophys. Res.*, 112, C08015, doi:10.1029/2006JC003729, 2007.
- 10 Sokal, R. R., and Rohlf, F. J. (Eds): *Biometry: the principles and practice of statistics in biological research*, WH Freeman and Co., New York, USA, 1995.
- Sørensen, H. L., Thamdrup, B., Jeppesen, E., Rysgaard, S. and Glud, R. N.: Nutrient availability limits biological production in Arctic sea ice melt ponds, *Polar Biol.*, 40(8), 1593–1606, doi:10.1007/s00300-017-2082-7, 2017.
- Spiese, C. E., Kieber, D. J., Nomura, C. T., and Kiene, R. P.: Reduction of dimethylsulfoxide to dimethylsulfide by marine phytoplankton, *Limnol. and Oceanogr.*, 54(2), 560-570, 2009.
- 15 Staley, J. T., and Gosink, J. J.: Poles apart: biodiversity and biogeography of sea ice bacteria, *Annu. Rev. Microbiol.*, 53(1), 189-215, 1999.
- Stefels, J.: Physiological aspects of the production and conversion of DMSP in marine algae and higher plants, *J. Sea Res.*, 43(3), 183-197, doi:10.1016/S1385-1101(00)00030-7, 2000.
- 20 Stefels, J., Steinke, M., Turner, S., Malin, G., and Belviso, S.: Environmental constraints on the production and removal of the climatically active gas dimethylsulphide (DMS) and implications for ecosystem modelling, *Biogeochemistry*, 83(1-3), 245-275, doi: 10.1007/s10533-007-9091-5, 2007.
- Stohl, A.: Characteristics of atmospheric transport into the Arctic troposphere, *J. Geophys. Res.-Atmos.*, 111, D11306, doi:10.1029/2005JD006888, 2006.
- 25 Stroeve, J. C., Markus, T., Boisvert, L., Miller, J., and Barrett, A.: Changes in Arctic melt season and implications for sea ice loss, *Geophys. Res. Lett.*, 41(4), 1216-1225, doi: 10.1002/2013GL058951, 2014.
- Sunda, W. K. D. J., Kieber, D. J., Kiene, R. P., and Huntsman, S.: An antioxidant function for DMSP and DMS in marine algae, *Nature*, 418(6895), 317-320, 2002.
- Thayer, G. W.: Comparison of two storage methods for the analysis of nitrogen and phosphorus fractions in estuarine water. *Chesapeake Sci.* 11, 155–158, 1970.

- Thomas, D. N. and Dieckmann, G. (Eds.): *Sea Ice* (2nd edn.), Wiley-Blackwell, 2010.
- Thomas, M. A., Suntharalingam, P., Pozzoli, L., Rast, S., Devasthale, A., Kloster, S., et al.: Quantification of DMS aerosol-cloud-climate interactions using the ECHAM5-HAMMOZ model in a current climate scenario, *Atmospheric Chemistry and Physics*, 10(15), 7425-7438, 2010.
- 5 Thronsdon, J., Hasle, G. R., and Tangen, K. (Eds.): *Phytoplankton of Norwegian coastal waters*, Almatier Forlag AS, Oslo, Norway, 2003.
- Tison, J. L., Brabant, F., Dumont, I., and Stefels, J.: High-resolution dimethyl sulfide and dimethylsulphonioacetate time series profiles in decaying summer first-year sea ice at Ice Station Polarstern, western Weddell Sea, Antarctica, *Journal of Geophysical Research: Biogeosciences*, 115(G4), 2010.
- 10 Tomas, C. R., and Hasle, G. R. (Eds): *Identifying marine phytoplankton*, Academic Press, New York, USA, 1997.
- Toole, D. A., and Siegel, D. A.: Light-driven cycling of dimethylsulfide (DMS) in the Sargasso Sea: Closing the loop, *Geophys. Res. Lett.*, 31(9), doi:10.1029/2004GL01958, 2004.
- Trevena, A. J., Jones, G. B., Wright, S. W., and Van den Enden, R. L.: Profiles of dimethylsulphonioacetate (DMSP), algal pigments, nutrients, and salinity in the fast ice of Prydz Bay, Antarctica, *Journal of Geophysical Research: Oceans*, 108(C5), 2003.
- 15 Trevena, A. J., and Jones, G. B.: Dimethylsulphide and dimethylsulphonioacetate in Antarctic sea ice and their release during sea ice melting, *Marine Chemistry*, 98(2-4), 210-222, 2006.
- Tschudi, M. A., Maslanik, J. A., and Perovich, D. K.: Derivation of melt pond coverage on Arctic sea ice using MODIS observations, *Remote Sensing of Environment*, 112(5), 2605-2614, 2008.
- 20 Turner, S. M., Nightingale, P. D., Broadgate, W., and Liss, P. S.: The distribution of dimethyl sulphide and dimethylsulphonioacetate in Antarctic waters and sea ice, *Deep Sea Research Part II: Topical Studies in Oceanography*, 42(4-5), 1059-1080, 1995.
- Twomey, S.: Pollution and the planetary albedo, *Atmos. Environ*, 8(12), 1251-1256, doi:10.1016/0004-6981(74)90004-3, 1974.
- 25 Van Bergeijk, S. A., and Stal, L. J.: The role of oxygenic phototrophic microorganisms in production and conversion of dimethylsulphonioacetate and dimethylsulfide in microbial mats, In *Biological and Environmental Chemistry of DMSP and Related Sulfonium Compounds* (pp. 369-379), Springer, Boston, MA, 1996.
- Van Bergeijk, S. A., Van der Zee, C., and Stal, L. J.: Uptake and excretion of dimethylsulphonioacetate is driven by salinity changes in the marine benthic diatom *Cylindrotheca closterium*, *Eur. J. Phycol.*, 38(4), 341-349, doi:10.1080/09670260310001612600, 2003.
- 30



- Vancoppenolle, M., Bitz, C. M. and Fichefet, T., Summer landfast sea ice desalination at point barrow: Model and observations, *J. Geophys. Res.*, 112, C04022, doi:10.1029/2006JC003493, 2007.
- Vancoppenolle, M., Notz, D., Vivier, F., Tison, J., Delille, B., Carnat, G., et al.: On the use of the mushy-layer Rayleigh number for the interpretation of sea-ice-core data, *The Cryosphere Discussions*, 7(4), 3209-3230, 2013.
- 5 Van Rijssel, M., and Gieskes, W. W. : Temperature, light, and the dimethylsulfoniopropionate (DMSP) content of *Emiliana huxleyi* (Prymnesiophyceae), *J. Sea Res.*, 48(1), 17-27, 2002.
- Vila-Costa, M., Simó, R., Harada, H., Gasol, J. M., Slezak, D., and Kiene, R. P., Dimethylsulfoniopropionate uptake by marine phytoplankton, *Science*, 314(5799), 652-654, doi: 10.1126/science.1131043, 2006.
- 10 Vogt, C., Rabenstein, A., Rethmeier, J., and Fischer, U.: Dimethyl sulphoxide reduction with reduced sulphur compounds as electron donors by anoxygenic phototrophic bacteria, *Microbiology*, 143(3), 767-773, 1997.
- Wait, B. R., Webster-Brown, J. G., Brown, K. L., Healy, M., and Hawes, I.: Chemistry and stratification of Antarctic meltwater ponds I: coastal ponds near Bratina Island, McMurdo Ice Shelf. *Antarctic Science*, 18(4), 515-524, 2006.
- Welsh, D. T.: Ecological significance of compatible solute accumulation by micro-organisms: from single cells to global climate, *FEMS Microbiol. Rev.*, 24(3), 263-290, 2000.
- 15 Weeks, W. F., and Ackley, S. F.: The growth, structure, and properties of sea ice, In *The geophysics of sea ice* (pp. 9-164), Springer, Boston, MA, 1986.
- Widell, K., Fer, I. and Haugan, P. M.: Salt release from warming sea ice, *Geophys. Res. Lett.*, 33(L12501), doi:10.1029/2006GL026262, 2006.
- 20 Willis, M. D., Burkart, J., Thomas, J. L., Köllner, F., Schneider, J., Bozem, H., Hoor, P. M., Aliabadi, A. A., Schulz, H., Herber, A. B., Leaitch, W. R. and Abbatt, J. P. D.: Growth of nucleation mode particles in the summertime Arctic: a case study, *Atmos. Chem. Phys.*, 16(12), 7663–7679, doi:10.5194/acp-16-7663-2016, 2016.
- Wolfe, G. V., Levasseur, M., Cantin, G., and Michaud, S.: Microbial consumption and production of dimethyl sulfide (DMS) in the Labrador Sea, *Aquat. Microb. Ecol.*, 18(2), 197-205, doi:10.3354/ame018197, 1999.
- 25 Woodhouse, M. T., Carslaw, K. S., Mann, G. W., Vallina, S. M., Vogt, M., Halloran, P. R., and Boucher, O. : Low sensitivity of cloud condensation nuclei to changes in the sea-air flux of dimethyl-sulphide, *Atmospheric Chemistry and Physics*, 10(16), 7545-7559, 2010.
- Worster, M. G., and Kerr, R. C.: The transient behaviour of alloys solidified from below prior to the formation of chimneys, *Journal of Fluid Mechanics*, 269, 23-44, 1994.

Wright, N. C. and Polashenski, C. M.: Open Source Algorithm for Detecting Sea Ice Surface Features in High Resolution Optical Imagery, Manuscript under review for the journal *The Cryosphere Discuss.*, doi:10.5194/TC-2017-154.

Yackel, J. J., Barber, D. G., and Hanesiak, J. M.: Melt Ponds on Sea Ice in the Canadian Arctic Archipelago: Part 1. Variability in morphological and radiative properties, *Journal of Geophysical Research*, 105(C9), 22049-22060, 2000.

5

Zemmelink, H. J., Gieskes, W. W. C., Klaassen, W., De Groot, H. W., De Baar, H. J. W., Dacey, J. W. H., Hintsa E.J., and McGillis, W. R.: Simultaneous use of relaxed eddy accumulation and gradient flux techniques for the measurement of sea-to-air exchange of dimethyl sulphide, *Atmos. Environ.*, 36(36), 5709-5717, doi:10.1016/S1352-2310(02)00577-0, 2002.

10 Zemmelink, H. J., Dacey, J. W., Houghton, L., Hintsa, E. J., and Liss, P. S.: Dimethylsulfide emissions over the multi-year ice of the western Weddell Sea, *Geophys. Res. Lett.*, 35(6), 2008.

Zhou, J. et al.: Physical and biogeochemical properties in land-fast sea ice (Barrow, Alaska): Insights on brine and gas dynamics across seasons, *Journal of Geophysical Research*, 118(6), 3172-3189, 2013.

15

20

## Figure captions

5 Figure 1: (a) Regional map showing the location of the four sampling stations (Ice1 to Ice4) (red circles) during the NETCARE/ArcticNet 2014 campaign. (b) MODIS imagery above the four sampling station (red circles) showing the ice conditions on 18 July 2014 in the sampling area. (c) Left to right, pictures of stations Ice1, Ice2, Ice3 and Ice4 with size scale. MPF stands for the Melt Ponds Fraction visually estimated from the bridge for stations Ice1, Ice2, Ice3 and Ice4.

10 Figure 2: Relationship between the concentrations of fresh DMS samples measured on board the ship via gas chromatography during the campaign and the concentrations of the corresponding preserved duplicate samples measured via coupled gas chromatography and mass spectrometry in a laboratory setting. The concentrations of the preserved DMS samples plotted are the sum of the three isotopes of DMS investigated in this study (m/z of 62, 63, and 68; see Materials and Methods).

15 Figure 3: In situ temperature (●) and bulk ice salinity (○) profiles of the sea ice surrounding the melt ponds sampled at stations Ice1 (a), Ice3 (b) and Ice4 (c). Temperature and salinity values of each 0.1 m sea ice section were used to calculate brine volumes (■), an indicator of sea ice permeability, throughout the full depth of sea ice (Cox and Weeks 1983, Petrich and Eicken 2010).

20 Figure 4: Temporal variations in DMSP<sub>d</sub> (a, c), and DMS (b, d) concentrations during the Ice1-MP1 and Ice4-MP1 incubation experiments. Both Light (○) and Dark (●) Treatments were initially amended with 100 nmol l<sup>-1</sup> of both D6-DMSP and <sup>13</sup>C-DMSO. Control Treatments (Δ) mimic natural concentration changes over time. In (a) and (c), vertical bars represent standard errors of mean values between duplicate samples.

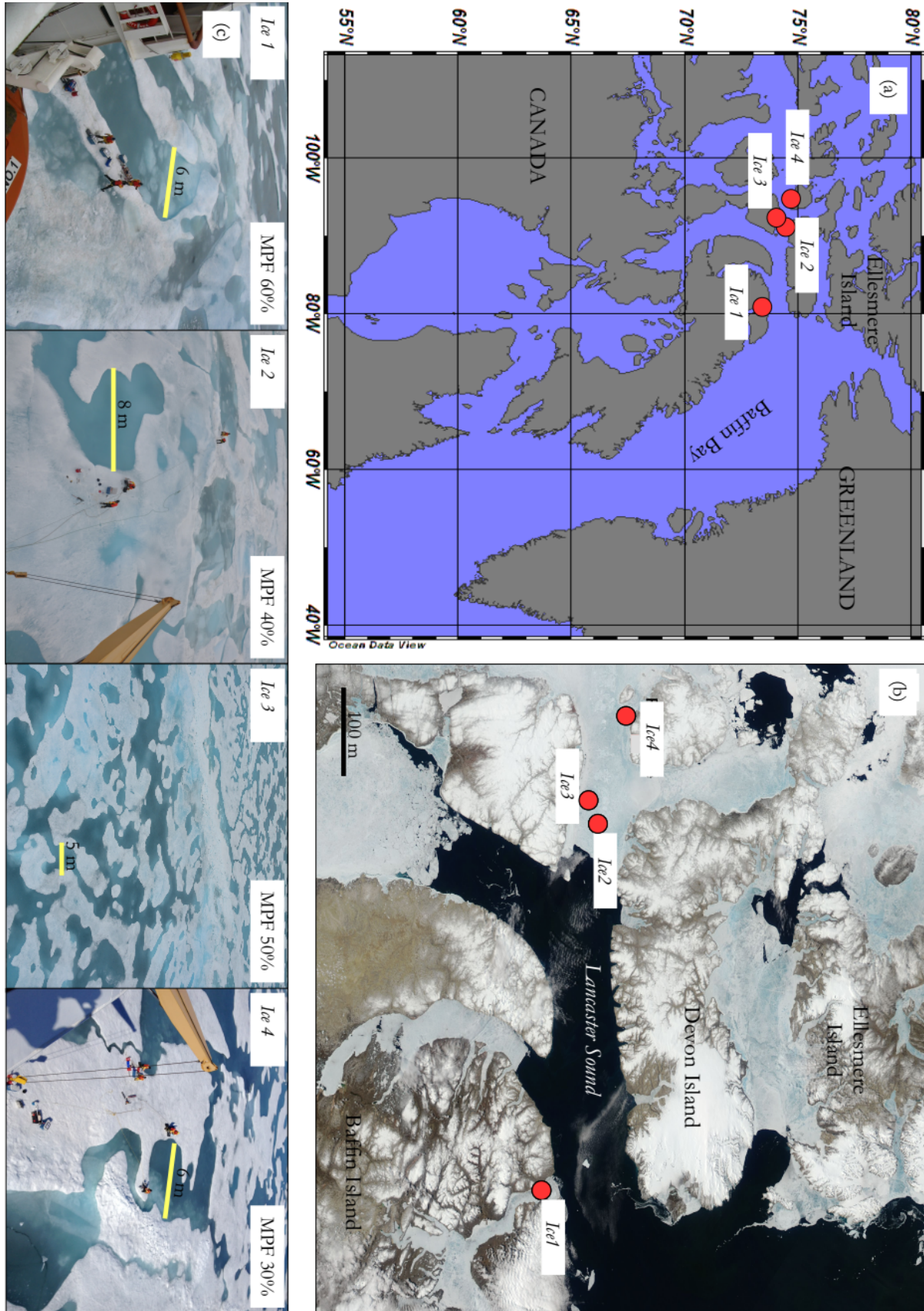
25

30

35

Figure 1: (a) Regional map showing the location of the four sampling stations (Ice1 to Ice4) (red circles) during the NETCARE/ArcticNet 2014 campaign. (b) MODIS imagery above the four sampling station (red circles) showing the ice conditions on 18 July 2014 in the sampling area. (c) Left to right, pictures of stations Ice1, Ice2, Ice3 and Ice4 with size scale. MPF stands for the Melt Ponds Fraction visually estimated from the bridge for stations Ice1, Ice2, Ice3 and Ice4.

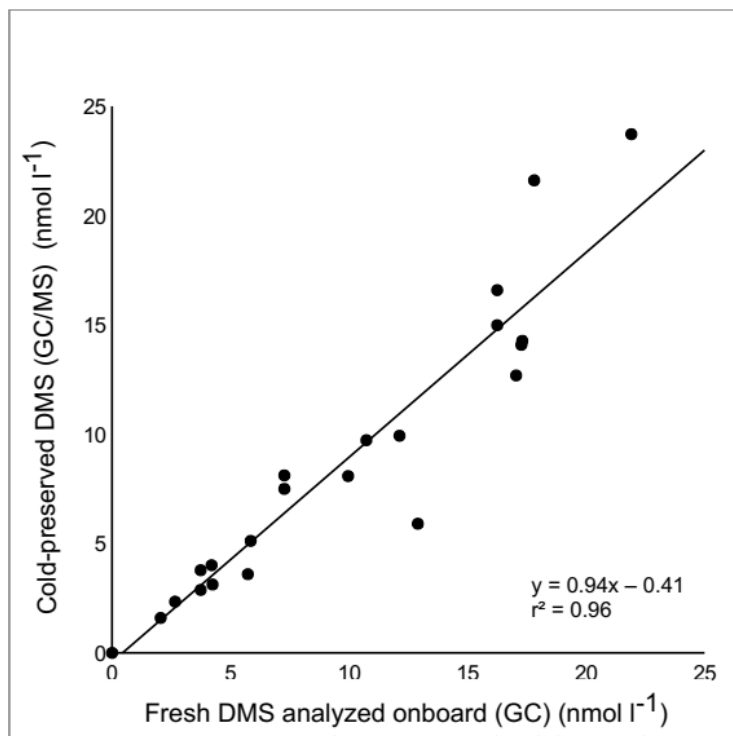
5



5

10

15

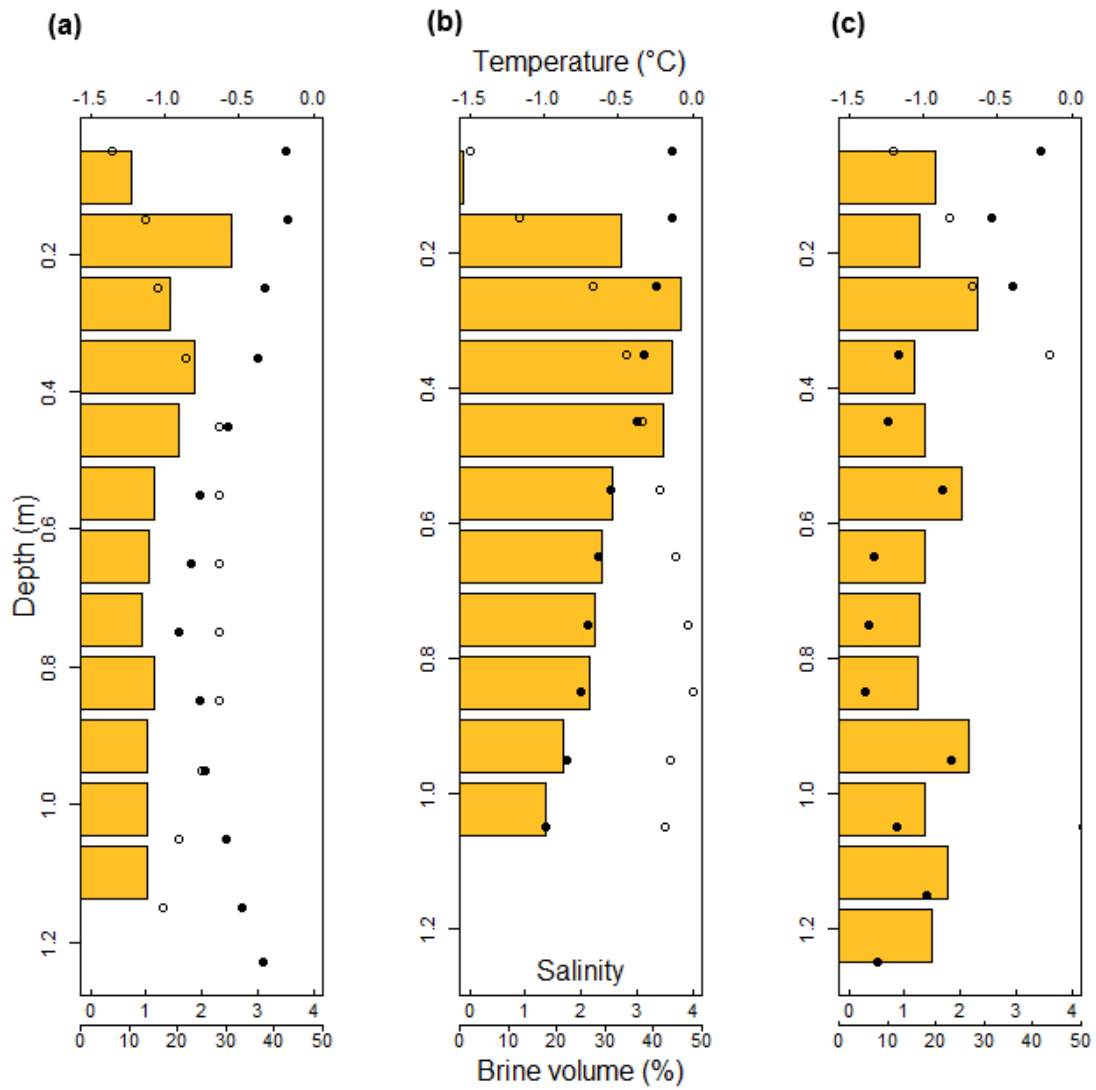


20

25

30

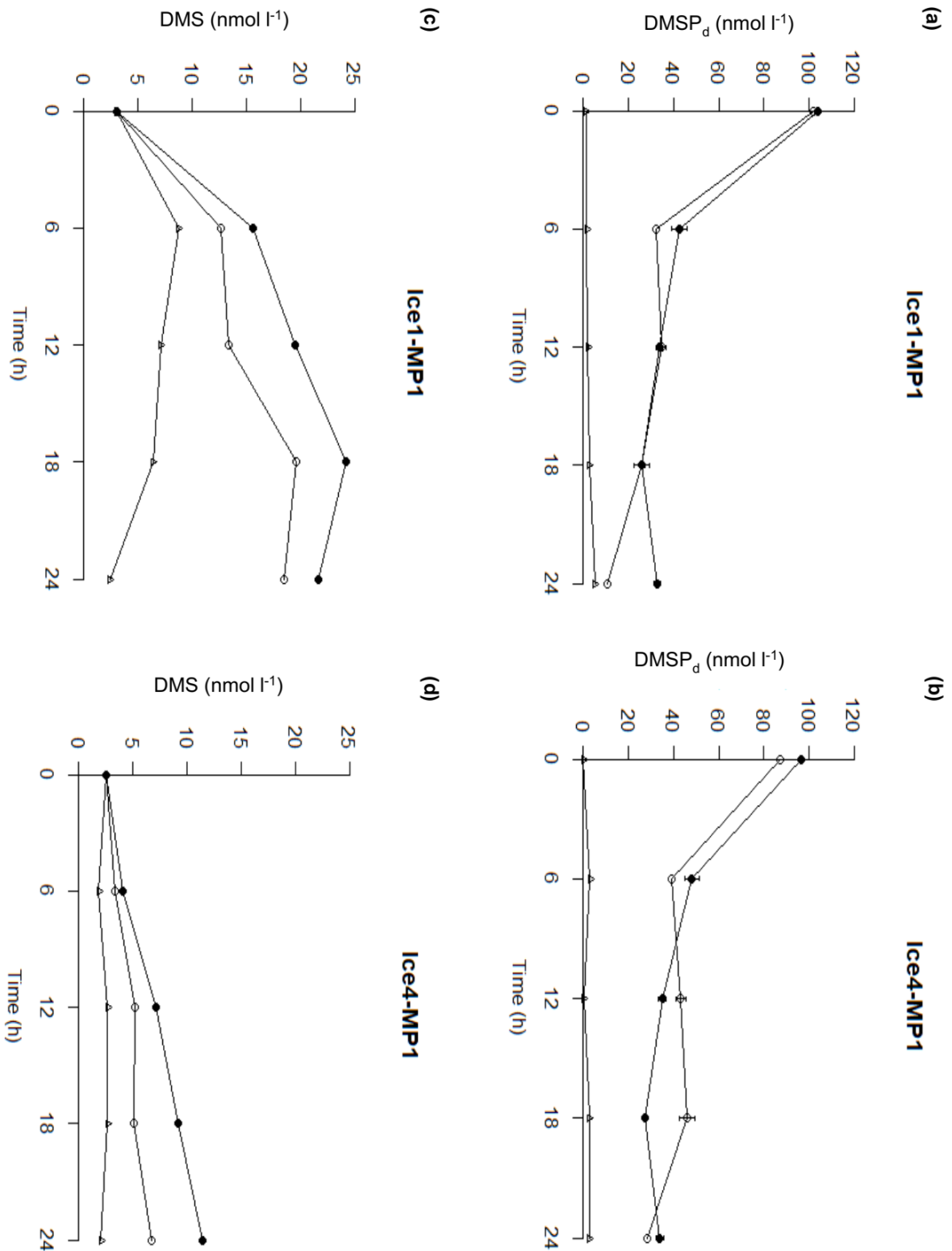
Figure 2: Relationship between the concentrations of fresh DMS samples measured on board the ship via gas chromatography during the campaign and the concentrations of the corresponding preserved duplicate samples measured via coupled gas chromatography and mass spectrometry in a laboratory setting. The concentrations of the preserved DMS samples plotted are the sum of the three isotopes of DMS investigated in this study ( $m/z$  of 62, 63, and 68; see Materials and Methods).



5

**Figure 3 (New figure added to the manuscript):** In situ temperature (●) and bulk ice salinity (○) profiles of the sea ice surrounding the melt ponds sampled at stations Ice1 (a), Ice3 (b) and Ice4 (c). Temperature and salinity values of each 0.1 m sea ice section were used to calculate brine volumes (orange bars), an indicator of sea ice permeability, throughout the full depth of sea ice (Cox and Weeks 1983, Petrich and Eicken 2010).

10



**Figure 4:** Temporal variations in DMSP<sub>d</sub> (a, b), and DMS (c, d) concentrations during the Ice1-MP1 and Ice4-MP1 incubation experiments. Both Light (○) and Dark (●) Treatments were initially amended with 100 nmol l<sup>-1</sup> of both D6-DMSP and <sup>13</sup>C-DMSO. Control Treatments (△) mimic natural concentration changes over time. In (a) and (b), vertical bars represent standard errors of mean values between duplicate samples. (Numbering order was modified as requested).

5

(New) Table 1: Physical characteristics of the sea ice surrounding the melt ponds. Note that only melt pond sampling (i.e. no ice sampling) was conducted at station Ice2 due to ship-related logistical constraints. A negative freeboard height indicates that the ice surface was locally below the mean sea level. n/a stands for non-available data. Ice thickness and freeboard values are averages of 7 (Ice1) to 8 (Ice3 and Ice4) ice cores sampled at each station.

5

Station	Sampling date	Snow and frozen snow* depth (cm)	Ice thickness (cm)	Freeboard (cm)
Ice1	Jul 18, 2014	0	121 ± 2	-1 ± 1
Ice2	Jul 20, 2014	0	n/a	n/a
Ice3	Jul 21, 2014	0 + 7*	113 ± 7	10 ± 2
Ice4	Jul 23, 2014	0	127 ± 1	7 ± 4

(Previously submitted Table 1: Physical characteristics of the sea ice surrounding the melt ponds. Note that only melt pond sampling (i.e. no ice sampling) was conducted at station Ice2 due to ship-related logistical constraints. A negative freeboard height indicates that the ice surface was locally below the mean sea level. n/a stands for non-available data.)

10

Station	Sampling date	Snow and frozen snow* depth (cm)	Ice thickness (cm)	Freeboard (cm)	Melt pond coverage estimate (%)
Ice1	Jul 18, 2014	0	121 ± 2	-1 ± 1	60
Ice2	Jul 20, 2014	0	n/a	n/a	40
Ice3	Jul 21, 2014	0 + 7*	113 ± 7	10 ± 2	50
Ice4	Jul 23, 2014	0	127 ± 1	7 ± 4	30

15

20

25



**Table 2: Physical, chemical and biological characteristics of the melt pond water. For melt pond depth, mean  $\pm$  standard deviation values are presented.**

Station	Melt pond #	Melt pond depth (m)	Melt pond salinity (psu)	Melt pond temperature (°C)
Ice1	MP1	0.18 $\pm$ 0.01	5.2	1.9
Ice1	MP2	0.18 $\pm$ 0.04	4.1	1.8
Ice2	MP1	0.29 $\pm$ 0.05	0.7	0.4
Ice2	MP2	0.19 $\pm$ 0.03	0.4	0.3
Ice2	MP3	0.12 $\pm$ 0.01	0.2	0.2
Ice3	MP1	0.07 $\pm$ 0.01	1.1	0.2
Ice3	MP2	0.10 $\pm$ 0.00	0.9	0.2
Ice4	MP1	0.12 $\pm$ 0.01	8.1	0.2
Ice4	MP2	0.11 $\pm$ 0.02	8.5	0.3

10

15

20

25

30

**Table 3: Reduced sulfur compound concentrations measured in situ in the melt ponds and the associated biological characteristics (abundance of high nucleic acid (HNA) bacteria, Chl *a* concentrations, and relative abundances of major taxonomic groups) of the melt pond water.**

Station	Melt pond	<i>In situ</i> DMSPp (nmol l <sup>-1</sup> )	<i>In situ</i> DMSPd (nmol l <sup>-1</sup> )	<i>In situ</i> DMS (nmol l <sup>-1</sup> )	Abundance of bacteria (HNA) (x 10 <sup>9</sup> cells l <sup>-1</sup> )	Chl <i>a</i> (µg l <sup>-1</sup> )	Abundance of algae (x 10 <sup>6</sup> cells l <sup>-1</sup> )	Dominant algal group
Ice1	MP1	2.2	1.3	3.0	0.24	0.48	2.00	Unidentified flagellates (50 %) Prasinophytes (ca. 25%)
	MP2	2.0	1.4	3.1		0.40		Unidentified flagellates (55 %) Prasinophytes (ca. 25 %)
Ice2	MP1	1.8	d.l.	d.l.	0.04	0.03	0.50	Unidentified flagellates (90 %) Pennate diatoms (ca. 28 %)
	MP2	2.4	d.l.	d.l.		0.09		Unidentified flagellates (50 %) Pennate diatoms (ca. 28 %)
	MP3	2.3	d.l.	d.l.		0.06		Unidentified flagellates (70 %) Pennate diatoms (ca. 28 %)
Ice3	MP1	2.0	d.l.	d.l.	0.02	0.05	0.30	Unidentified flagellates (45 %) Chrysophytes (29 %)
	MP2	2.3	d.l.	d.l.		0.04		Unidentified flagellates (55 %) Chrysophytes (23 %)
Ice4	MP1	4.0	d.l.	2.6	0.15	0.18	1.00	Unidentified flagellates (50 %) Pennate diatoms (20 %)
	MP2	3.7	1.1	6.1		0.20		Unidentified flagellates (60%) Pennate diatoms (25 %)

10

15

20

25

**Table 4: Spearman's rank correlation coefficients between key in situ variables measured in the melt ponds. \* indicates a 0.05 significance level.**

	DMS	Salinity	Temperature	Chl <i>a</i>
DMS		0.84*	0.51	0.84*
Salinity			0.40	0.56
Temperature				0.60

10

15

20

25

30

35

Table 5: In situ DMSP<sub>p</sub>, DMSP<sub>d</sub> and DMS change rates measured during the incubation experiments conducted in melt ponds Ice1-MP1 and Ice4-MP1. Hourly rates for DMSP<sub>d</sub> and DMS net changes measured between T<sub>0</sub> and T<sub>6</sub> as well as T<sub>6</sub> and T<sub>24</sub> are derived from the slope of DMSP<sub>d</sub> and DMS concentrations vs. time, respectively. Daily DMSP<sub>d</sub> change rates are calculated as the difference between the DMSP<sub>d</sub> concentrations measured at T<sub>24</sub> and T<sub>0</sub>. Daily DMS change rates are calculated as the difference between the DMS concentrations measured at T<sub>24</sub> and T<sub>0</sub>. Rates measured over the first 6 h and between T<sub>6</sub> and T<sub>24</sub> are expressed in nmol l<sup>-1</sup> h<sup>-1</sup>. Other rates are expressed in nmol l<sup>-1</sup> d<sup>-1</sup>.

10

Station	In situ DMSP <sub>p</sub> change rates	In situ DMSP <sub>d</sub> change rates		In situ DMS change rates	
	(nmol l <sup>-1</sup> d <sup>-1</sup> )	(nmol l <sup>-1</sup> h <sup>-1</sup> ) (6h)	(nmol l <sup>-1</sup> d <sup>-1</sup> )	(nmol l <sup>-1</sup> h <sup>-1</sup> ) (6h)	(nmol l <sup>-1</sup> d <sup>-1</sup> )
Ice1-MP1	-2.2	0.0	4.0	1.0	1.2
Ice 4-MP1	-1.9	0.0	-0.1	-0.1	0.0

15

20

25

30

35

Table 6: Potential net DMSP<sub>d</sub> change rates, potential net DMS change rates and light-associated DMS sinks measured during the incubation experiments conducted in melt ponds Ice1-MP1 and Ice4-MP1. Clear and shaded horizontal lines regroup the rates measured under natural light (L-DMSP/O) and in the dark (D-DMSP/O), respectively. Hourly rates for potential DMSP<sub>d</sub> and DMS net changes between T<sub>0</sub> and T<sub>6</sub> as well as T<sub>6</sub> and T<sub>24</sub> are derived from the slope of DMSP<sub>d</sub> and DMS concentrations vs. time, respectively. Daily potential net DMSP<sub>d</sub> change rates are calculated as the difference between the DMSP<sub>d</sub> concentrations measured at T<sub>24</sub> and T<sub>0</sub>. Daily potential net DMS change rates are calculated as the difference between the DMS concentrations measured at T<sub>24</sub> and T<sub>0</sub>. Rates of light-associated DMS sink were measured as the difference of DMS accumulation between L-DMSP/O and D-DMSP/O after the 24h incubation. Rates measured over the first 6 h and between T<sub>6</sub> and T<sub>24</sub> are expressed in nmol l<sup>-1</sup> h<sup>-1</sup>. Other rates are expressed in nmol l<sup>-1</sup> d<sup>-1</sup>.

10

Station	Potential net DMSP <sub>d</sub> change rates			Potential net DMS change rates		Light-associated DMS sinks
	(nmol l <sup>-1</sup> h <sup>-1</sup> ) (T <sub>0</sub> -T <sub>6</sub> )	(nmol l <sup>-1</sup> h <sup>-1</sup> ) (T <sub>6</sub> -T <sub>24</sub> )	(nmol l <sup>-1</sup> d <sup>-1</sup> )	(nmol l <sup>-1</sup> h <sup>-1</sup> ) (T <sub>0</sub> -T <sub>6</sub> )	(nmol l <sup>-1</sup> d <sup>-1</sup> )	(nmol l <sup>-1</sup> d <sup>-1</sup> )
Ice1-MP1	-11.6	-1.2	-91.5	1.6	15.4	3.2
	-10.2	-0.6	-71.3	2.1	18.6	---
Ice 4-MP1	-8.1	-0.5	-59.2	0.1	4.2	4.7
	-8.1	-0.9	-62.6	0.3	8.9	---

10

15

20

5 **Table 7: (m/z 62) and (m/z 68) DMS concentrations after 24h incubation in the Control, L-DMSP/O and D-DMSP/O Treatments. Relative contribution (%) of natural DMS and D6-DMSP to the total DMS measured at T<sub>24</sub> in the Control, L-DMSP/O and D-DMSP/O Treatments during the incubation experiments with water from Ice1-MP1 and Ice4-MP1. Natural DMS signature = (m/z 62); signature of DMS derived from D6-DMSP = (m/z 68). No (m/z 63), which represents the signature of DMS derived from <sup>13</sup>C-DMSO, was retrieved either after 12 h (not shown) or 24 h.**

Incubation	Treatment	(m/z 62) DMS (nmol l <sup>-1</sup> )	(m/z 68) DMS (nmol l <sup>-1</sup> )	(m/z 62) % of total DMS	(m/z 68) % of total DMS
Ice1-MP1	Control	3.0	0.0	100	0
	L-DMSP/O	4.1	14.4	22	78
	D-DMSP/O	6.6	18.2	27	73
Ice4-MP1	Control	2.3	0.0	100	0
	L-DMSP/O	1.3	5.1	20	80
	D-DMSP/O	4.2	7.9	35	65