

Interactive comment on “Dimethylsulfide dynamics in first-year sea ice melt ponds in the Canadian Arctic Archipelago” by Margaux Gourdal et al.

G. Carnat (Referee)

gauthier.carnat@gmail.com

Received and published: 2 February 2018

General comments: 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

C1

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. 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. Specific comments: Please find a list of suggested references and reading at the end of the review. P2, L2 (and throughout the manuscript): “first-year” instead of “first year”. P2, L3 (and throughout the manuscript): sea ice instead of sea-ice. Please be consistent throughout the manuscript. P2, L6: “In the Eastern Canadian Arctic”, I would use “Canadian Arctic Archipelago” to be consistent with the title. 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. P2, L9: “Experiments conducted with” rather than “Results from experiments”. This is a

C2

little bit redundant with the next sentence. P2, L10: Bracket missing here. 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 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. P3, L12: "DMS-derived sulfate aerosols". P3, L14: Please indicate the two different backscattering effects of DMS-derived sulfate aerosols (direct and indirect through CCN). 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 sentence and talk about the influence of DMS on a more regional scale. P3, L16: "In remote pristine marine areas such as the polar regions". P3, L16: "Could be particularly important". P3, L19: Please add a reference here. P3, L20: The study of Rempillo et al. (2011) could also be cited here. 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. P3, L24: The reference is not correct. It should be Green and Hatton (2014). P3, L23-24: "Cellular metabolite" rather than "cellular compound", compound is a little bit vague. 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. P3, L32: in-situ. P3, L33: It would be nice to indicate in a short sentence how DMSP is released from the cell. 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

C3

suggest to rewrite following these lines: ". . . found in several phytoplankton species (DMSP particulate, or DMSPp) (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 DMSPd) where it is readily used by heterotrophic bacteria as carbon and sulfur sources (Kiene et al., 2000; Simo, 2001; Vila-Costa et al., 2006). The fraction of DMSPd 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 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; 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; Simo 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 (Simo and Pedros-Alio, 1999a)." P4, L5: It would be nice to write one or two sentences on particulate DMSO. P4, L10: As explained in my general comments, I think you need here

C4

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. Also, a few sentences on the specificity of the mobility of compounds within sea ice (i.e. permeability) should appear in this paragraph as it is a key part of your study. 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. 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. 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 difference in microbial community development between the Arctic and Antarctic. 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. 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. 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. P4, L25: You

C5

could already indicate here between brackets (logistical constraints) why basic physical measurements were not conducted at Ice2. P4, L26: Please already define freeboard here. 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. 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. 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. . .". P4, L30: (and throughout the manuscript). Check for spacing between 5 and cm. I do not know what the recommendations of Biogeosciences are. 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. P4, L32: "the bulk salinity of the melt aliquot". P4, L32: Permeability to fluid/gas transport is a more appropriate term than porosity here. 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 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°C for an ice salinity of 5. . .not that the ice is permeable when the ice temperature is warmer than -5°C and the salinity higher than 5. P5, L7: Additional details are needed here.

C6

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 location in Fig1. P5, L11: How many replicates? It is not clear if chl a was measured on the ship or the filters stored. P5, L23-24: This is slightly confusing. Stored in liquid nitrogen (-196°C) or kept frozen at -80°C? 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. P5, L30: "to fill the glass serum bottles" remove the "the". P6, L11: Consider cutting in two sentences. "...into 5 ml Falcon™ tube. DMSPd was quantified...". 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. P6, L16: Dacey and Blough (1987) is perhaps a better reference here than Levasseur et al. (2006). P6, L26: "fresh water", do you mean milliQ water? Please specify. P6, L30: Consider using "duplicate" instead of "duplicated". 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. P8, L5: Is any fractionation expected during storage? P8, L6-10: Please provide the overall precision of the methods. P8, L9-10: This is very nice to read. P9, L5: Please add this 5 m information in the section 2.1 of the materials and methods part. 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. P9, L29: Please replace (see discussion) by "This will be discussed in section...". P11, L11: The use of "significantly" implies a statistical test which is not provided. 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). P12, L1-2: See my previous comment. Please read the study of Leck and Persson (1996), cited in Levasseur (2013). There is also some

C7

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. P12, L5: As stated before, I think this sentence is misleading and should be remove 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. 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. P12, L17 and 23: "Sea spray". 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). 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. 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-

C8

work leading to the process called flushing. This further decreases the bulk ice salinity down to values way under 2 psu 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. 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 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 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 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. P14,

C9

L6-8: "over-flooding of sea ice". Replace by "flooding of the ice surface". Over-flooding is an odd term. P14, L6: Flooding could be better defined. P16, L7: Again, consider other data sets available. P16, L16: Modify "over-flooding". 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 towards the atmosphere. P16, L24: These numbers should be put in perspective. How do they compare to the sea ice, ocean reservoirs? P16, L26: Is the average depth calculated from your data set or from literature observations? Your data set is relatively small. P16, L29: Wind velocity but also a better understanding of gas exchange between small fetch melt ponds and the atmosphere. References: Check the alphabetic order, Giamarelou et al. should be after Garrison. Figures and tables: 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. Table 7: Please be consistent with the significant digits. 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. Figure 3: Odd lettering of the figures.

Suggested references to read and/or add: -Quinn and Bates (2011). The case against climate regulation via oceanic phytoplankton sulphur emissions. *Nature*. -Green and Hatton (2014). 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*. -Rempillo et al. (2011). Dimethyl sulfide air-sea fluxes and biogenic sulfur as a source of new aerosols in the Arctic fall. *Journal of Geophysical Research*. -Lana et al. (2011). An updated climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean. *Global Biogeochemistry*. -Lyon et al. (2016). Role of dimethylsulfoniopropionate as an osmoprotectant following gradual salinity shifts in the sea-ice diatom *Fragilariopsis cylindrus*. *Environmental Chemistry*. -Karsten et al. (1996). Dimethylsulfoniopropionate production in phototrophic organisms and its physiological function as a cryoprotectant. *Biological and Environmental Chemistry of DMSP and Related Sulfonium Compounds*. -Levasseur (2013). Impact of arctic meltdown on

C10

the microbial cycling of sulphur. *Nature Geosciences*. -Leck and Persson (1996). The central Arctic Ocean as a source of dimethyl sulfide: seasonal variability in relation to biological activity. *Tellus B*. -Miller et al. (2015). Methods for biogeochemical studies of sea ice: The state of the art, caveats, and recommendations. *Elementa: Science of the Anthropocene*. -Lepparanta and Manninen (1988). The brine and gas content of sea ice with attention to low salinities and high temperatures. Finnish Institute of Marine Research Internal Report. -Carnat et al. (2013). 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*. -Carnat et al. (2014). 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*. -Zhou et al. (2013). Physical and biogeochemical properties in land-fast sea ice (Barrow, Alaska): Insights on brine and gas dynamics across seasons. *Journal of Geophysical Research*. -Dacey and Blough (1987). Hydroxide decomposition of dimethylsulfoniopropionate to form dimethylsulfide. *Geophysical Research Letters*. -De Mora et al. (1996). Aspects of the biogeochemistry of sulphur in glacial melt water ponds on the McMurdo Ice Shelf, Antarctica. *Antarctic Science*. -Cox and Weeks (1983). Equations for determining the gas and brine volumes in sea ice samples. *Journal of Glaciology*. -Nomura et al. (2012). Direct measurements of DMS flux from Antarctic fast sea ice to the atmosphere by a chamber technique. *Journal of Geophysical Research Oceans*.

Interactive comment on *Biogeosciences Discuss.*, <https://doi.org/10.5194/bg-2017-432>, 2017.