Interactive comment on "The effect of warm summer 2012 on seasonal and annual methane dynamics in adjacent small lakes on the ice-free margin of Greenland" by Sarah B. Cadieux et al.

Anonymous Referee #1 Received and published: 15 August 2016

*response from authors is in blue

Summary

The manuscript describes a study on dissolved CH4 concentration in five arctic shallow lakes located in Greenland. They used here five data sets (from summer 2012 to winter 2014) on the Southwest ice-free margin of Greenland. The aim of the study was focused on the effect of one high warming event occurred in summer 2012 on CH4 concentration profiles and compared it with subsequent years (2013 and 2014). The study of CH4 dynamics in lakes is a topic of broad scientific interest as lakes represent an important source of this gas to the atmosphere.

I recognize that it is a difficult task to study lakes in these extreme environments, and data coming from them are therefore valuable. The manuscript is not very clear in demonstrating how the warm 2012 summer influenced CH4 dynamics in these lakes. Even, this study shows minor effects of the 2012 warm summer on CH4 dynamics (showed in Figure 7), and it is very difficult to correlate the minor effects to any partic- ular phenomena (showed in Figure 8).

Likewise, the authors should always make clear when data have been previously published. I was surprised that several data in Tables, Figures (Figure 6 and 7) and Map (Figure 2) are the same (or at least very similar) than those reported in another manuscript from the same authors (Cadieux et al. 2016); and no reference is made to that previous study (and/or indicated in those tables and figures). I also want to point out, that there are strong similarities in the DOC and pH data presented in Table 1 and Table 2 (for DOC) for open-water conditions 2012 in this manuscript and data presented in Table 1 for open water conditions in 2013 from Cadieux et al. (2016).

The manuscript is well written, although some sections are not totally mature yet and therefore the manuscript lacks a clear focus and structure. I think that some of the analysis are speculative and/or over-interpreted and numerous issues in the method section must be better addressed.

First, we thank the reviewer for their positive comments on the paper and feel they have raised some valid concerns. The reviewer is correct in noticing that some data and parts of figures are in Cadieux et al. 2016. However, its important to note that in Cadieux et al. 2016, only 3 lakes (EVV Upper lake, Teardrop lake, and Potentilla lake) are discussed for only open water conditions in 2013 and ice covered conditions 2014. In order to be explicit about data that is previously published, we have revised tables and figures to cite which lakes and data points have been previously published.

The reviewer is also correct in noticing that in Table 1, these physico-chemical characteristics of the lakes in open-water conditions in 2012 are the same as in Cadieux et al. (2016) - which is inaccurately labeled water conditions for 2013 when they are 2012. For the paper here, we revised the table text to include that this data is also in Cadieux et al. 2016 and will take steps to revise the other paper so both match.

Specific comments

The introduction contains mixed statements related to temperature effects on CH4 production/oxidation/storage in the water column (e.g. temperature dependencies on CH4 production is described in two sentences in second and fourth paragraphs). I would recommend reorganizing the ideas to improve the introduction flow (which should go from general to specific).

Thank you for this suggestion to strengthen the introduction. In order to go from general to specific, as well as remove repetitive information, the organization now goes from discussion of lakes and climate, to methane production and consumption, to focus of this study.

Likewise, it is necessary to carefully review the literature to avoid controversial state- ments like the authors indicate at the end of the introduction "This work provides the first measurements of dissolved CH4 concentrations under both open-water and ice- covered conditions for consecutive years in small, Arctic lakes". From the literature that I know (and for sure I am missing a vast amount of studies), there are previous studies or multiyear dissolved CH4 concentrations in water column, in similar latitudes. Some of these previous works measured dissolved CH4 concentration through and over sev- eral years. I suggest some readings: Kaankala et al. (2006), Bellido et al. (2009), Karlsson et al. (2013), Greene et al. (2014), Miettinen et al. (2015), Tan et al. (2015), among others.

We appreciate the reviewer bringing this to our attention. Previously, in our literature search, we had not found Karlsson et al. (2013), Miettienen et al. 2015 and Greene et al. 2014 which all also describe both multiyear and multiseason results. The others described above are only for one season or one year. We have removed this sentence accordingly.

The description of the methods is the most important section to understand what the authors did. This section has to be improved substantially. Firstly, I found a number of cases in which devices or sample preparation are not full described (e.g. electronic submersible pump, total organic carbon analyzer, passive diffusion bags PDBs, HCl concentration, dilution correction for CH4 measurements). Secondly, littoral sediment CH4 bubble sampling method (used in this manuscript) is a very unspecific method. While in Cadieux et al. (2016) the method was used in combination with the isotopic analysis (isotopic values are helping to understand CH4 dynamics), in this manuscript, values of CH4 are given without determining the volume of sediment samples (as com- mented in the method section). Therefore, what is the point to include very speculative values of CH4 concentration from the littoral. Thirdly, I consider it would be necessary to

describe briefly the methods, even if they are previously described (Cadieux et al. 2016), to avoid excessive self-citation and tedious reading. Finally, the statistical anal- yses need to be clarified. Some of them does not make sense, as written, and specific information is required to understand how data analysis was made e.g., mean/median temperature and CH4, profile values, seasonal, sectional.

The reviewer makes good point that this paper would be strengthened by describing the methods in more detail. We revised the methods section in the following ways in an attempt to address the concerns noted above:

- Regarding the DOC measurement, a reference has been added to describe the questions of methodology (Oviedo-Vargas, D., Royer, T.V., Johnson, L.T., 2013. Dissolved organic carbon manipulation reveals coupled cycling of carbon, nitrogen, and phosphorus in a nitrogen-rich stream. Limnology and Oceanography 58, 1196-1206.).
- We specified the model and type of electronic submersible pump: "Water for chemical analysis was collected from the water column using a Narrow Diameter Supernova[™] electronic submersible pump."
- The methods for dissolved methane sampling have been briefly expanded: "With the exception of Potentilla lake under ice-covered conditions in 2014, water samples for dissolved CH_4 in the water column were collected using an electronic submersible pump. Samples were collected at 0.25-1.0 m intervals through the water column and were immediately stripped in the field using a headspaceequilibrium technique (Westendorp 1985) to extract CH₄ from water. At each depth interval, 500 mL of water was collected into a 1 L Erlenmeyer flask and vigorously shaken for 1 minute. Headspace gas in the flask was displaced into a Cali-5-Bond bag using surficial lake water (Cadieux et al., 2016). Under icecovered conditions in 2014, dissolved CH₄ in Potentilla lake was collected using a string of passive diffusion bags (PDBs) deployed in the lake for 5 days in order to obtain a high-resolution profile of dissolved CH_4 in the water column (Goldman et al., 2016). The PDBs are composed of a polyethylene membrane with a protective plastic mesh and are commercially available from EON Products Inc. (Georgia, USA). After 5 days, PDBs were retrieved from the lake and dissolved gas was sampled immediately in the field using the equilibrium gas stripping method described above. Further details regarding PDB methodology, preparation and applicability can be found in Goldman et al. (2016)."
- We acknowledge that the littoral methane concentrations are speculative, as we did not measure a concentration of sediment in order to normalize lake-to-lake.
 We have added a statement to explicitly state that this is just an estimation: "We were unable to quantify the volume of sediment samples, therefore concentrations of CH₄ in gas collected from littoral sediments cannot be

converted into pool size of CH_4 in the littoral sediments, and are only an approximation of CH_4 concentration."

- In order to not over-analyzed results, the following was added to the littoral CH₄ section: "However, it is important to note that littoral CH₄ concentrations are an estimate, as a volume of sediment/sample was unmeasured. Therefore, it is possible that the increase in littoral CH₄ concentrations is not the result of increased CH₄ production, but of a different amount of sediment disturbed."
- Statistical analysis section has been clarified as to what was tested and why: "Statistical analyses were made using IBM SPSS Statistics. Concentrations of CH₄ and chemical variables for all study lakes during each season were assessed for normal distribution via the Kolmogorov-Smirnov test, and were found to be nonnormally distributed. Student's t test of unequal variance was used for testing statistically significant differences in concentrations of CH₄ between open-water and ice-covered conditions, as well as from one year to another. Systematic changes in aquatic chemistry and CH₄ concentrations were analyzed using linear regression, in order to assess if CH₄ concentrations were related to variables such as DO, temperature, DOC and conductivity."

Through the results and discussion section some Figures are used to explain variations and significant differences between lakes. In data from Figure 7, it is impossible to note the range reported in surface waters and depth axes are missing in some sub-figures (making impossible to see clearly the depth profile). Moreover, in data analysis from Figure 8 (wrongly named Figure 7 in Page 9, Line 307), it is impossible to see when CH4 vs. DO and CH4 vs. T are related or not. Likewise, some discussion sections are not well focused on the main issue and over interpret results. Some examples are:

Regarding the figures, Line 307 was revised to Figure 8 and depth axes have been added to all of the panels in figure 7.

We agree with the reviewer that it is difficult to note the range of surface water CH₄ concentrations in Figure 7. In order to clarify this, a new table has been made that defines the surficial values under open-water conditions in all of the lakes:

Table 3: CH₄ concentrations (μ M) in surface waters under open-water conditions in 2012 and 2013.

	2012	2013
EVV Upper	2.7	0.9
EVV Lower	11.5	2.7
Teardrop	27.8	2.4
Potentilla	2.6	1.3
South Twin	4.3	3.5

We also agree with the reviewer that in Figure 8_it is difficult to see the trend. This is because there is not a very statistically significant trend between CH₄ concentration and

temperature/dissolved oxygen. This is why we had included r^2 values on each of the figures. We have gone through the text to ensure that we do not over analyze this non-significant trend.

i) competition for substrates favors sulfur reduction (SR) and methanogenesis typically does not occur until SO42- is exhausted and SR rates have decreased (Lovely & Klung 1983, Lovely & Klung 1986, Scholten et al., 2002, Ward & Winfrey 1985). However, EVV Upper lake did not have the lowest concentrations of CH4 in the water column, suggesting there was sufficient reduced carbon substrates to fuel both SR and methanogenesis. Therefore, while aquatic chemistry in the water 320 column may be a factor influencing CH4 production, it alone is insufficient to explain the variation in CH4 concentrations observed lake-to-lake, as well as seasonally and annually."

Give the small sample size for each lake, our statistical power is limited for aquatic chemistry. In keeping with the reviewer's suggestion, the last sentence has been revised to: "Therefore, while aquatic chemistry in the water column could be a factor influencing CH₄ production, at the level of this investigation, it alone is likely insufficient to explain the variation in CH₄ concentrations observed lake-to-lake, as well as seasonally and annually."

ii) all section "6.3 Effects of temperature on CH4", and

In keeping with the reviewer's suggestion, we have revised section 6.3 to explicitly mention that the relationships observed are at a specific date and time. For example: "Despite the absence of a strong linear relationship between water temperature and CH₄ concentrations, warmer ground-level air temperatures correspond with increased CH₄ both in the water column and the sediments in the study lakes at the time of sampling."

iii) you don't have thorough information on the ice phenology to indicate that "Our re- sults suggest that changes in the duration of seasonal ice cover will, in turn, result in changes in inventories of under-ice CH4. As the duration of ice cover decreases, the amount of CH4 stored under ice 455 cover will likely decrease due to the shorter time for accumulation, potentially reducing the amounts of CH4 emitted during ice-breakup and spring overturn.". I think, the results are not reliable to support such statements.

We agree with the reviewer that we don't have dates for ice-in or ice-out or other ice phenology information. However, with the data we do have, we can speculate what may happen to CH₄ inventories as ice-cover duration decreased. In keeping with the reviewer's comments, we revised this section accordingly: "Currently, the largest efflux of CH₄ from our study lakes occurs during ice-breakup and spring overturn. Changes in the duration of seasonal ice-cover will result in changes in inventories of under-ice CH₄. We predict that as the duration of ice cover decreases, the amount of CH₄ stored under ice cover will likely decrease due to the shorter time for accumulation. If the amount of stored CH₄ under ice-cover decreases, this will potentially reduce the amount of CH₄ emitted during ice-breakup and spring overturn."

References

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Tan, Z., Zhuang, Q., Water Anthony, K.: Modeling methane emissions from arc- tic lakes: Model development and site-level study, J. Adv. Model. Earth Syst., 07, doi:10.1002/2014MS000344., 2015. (note: for sure to develop the models, they used multi-year dissolved CH4 concentration data).

We thank the reviewer for including the full citations for the references suggested. These have been incorporated into the text.