COMMENTS TO THE EDITOR

Associate Editor Decision: Reconsider after major revisions (19 Jul 2017) by Gwenaël Abril
Comments to the Author:
Decision #3 on bg-2017-22 Methane distribution and oxidation around the Lena Delta in summer 2013

Dear Authors
I have received a second review of anonymous referee #3, and her/his evaluation of your revised MS was quite severe (see report). Note that this reviewer is an expert in the field. I made my own reading of your revised MS, including the modifications in the first version, and, indeed, I agree with referee#3 that your revisions were too superficial and not sufficient to satisfy all initial comments and reach the standards of a high quality journal like Biogeosciences. However, I believe this work must be published in BG, but only after a more careful and detailed revision. Critics concern:

(1) presentation of the MS and formatting of figures and tables, and legends;
The Ms has now been checked by a professional editing service; and the figures have been polished.

(2) a too superficial discussion that does not adequately refer to the literature in order to put the presented results in a broader context;
The discussion on methane concentrations and methane emission, as well as the conclusion have been completely rewritten. Relating the data also to worldwide data sets, and extrapolating our data to the warming of the Arctic climate.

(3) some imprecise or inappropriate statements in the interpretation of the data.
We checked the whole Ms again, as well as the editorial service, and think the text has much improved now.

I would be pleased if you could revise your MS in order to satisfy the referee's comments (and mine you will find below), and provide a detailed list of responses and a description of what as been changed in the MS.
Looking forward to reading this soon.
Best Regards
Gwenaël Abril, BG associate editor

Additional comments:
L204, what were the criteria used to delimit the two rectangular area. Appendix Fig. A1 is not necessary if the rectangles appear in Fig.1
We tried to envelope the whole study area, which was best obtained with two rectangles which are bordered by the most southern,northern, eastern and western stations. This *area is now indicated as dashed line in the map.*
The text is now changed to:
"Two rectangles which are bordered by the most southern, northern, eastern and western stations gave a good estimation of investigated area Figure 1."

L234 and throughout MS: "pale orange" is inappropriate as in fact it is "pale pink", pale orange appearing between the yellow and the red in the colour grid. All the figures drawn with ocean data view are of poor quality (coloured points are very large and sometimes overlap) and the mentions "@depth(m)=last" are not understandable. Please improve also the legends.
We have polished now all odv plots, including the legends. When some data points are above the range, their number is now indicated with their real number.

Notation „riverine water“ is not conventional and unacceptable
But the word is in the Oxford Dictionary, with" riverine = Relating to or situated on a river or riverbank; riparian". The professional editing service also had no objections……

I believe the information in the appendix tables 1 and 2 could appear together in a same table in the MS.
Ok I have incorporated them again into the text, even though referee 3 wanted these table in the supplement…..

Why didn't you test the correlation of CH4 concentration and MOX activity only after separating "river" "mixed" and "artic" and for the whole dataset?
Best results were obtained when splitting the dataset. And as riverine and polar water are separated by a strong pycnocline, we think it is more reasonable to do the statistics separately.
L 248 ff
"When applying our water masses (riverine, mixed and polar), we observed significantly different methane concentrations in these water masses, with medians of 22, 19 and 26 nmol L^{-1} ($p = 0.03$), respectively(table1). Therefore, the linear correlation analysis was performed separately for the different water masses.!

Please also consider a presentation of CH4 concentrations versus salinity, as this is very classical and broadly used in the literature.
A new figure 8 with methane versus salinity is now added to the ms and "we thus conclude that the classical way of river water dilution does not apply for the Lena Delta". Line 373 ff

L317 The question raised by the referee of the general low CH4 concentrations in artic rivers and estuaries compared to other regions in the world is to my opinion fundamental and deserves a specific paragraph in the discussion: why are CH4 concentrations lower in artic estuaries?
A additional paragraph, comparing tropical, temperate and arctic waters has been added, lines L516 ff

L344 Reader is lost here. CH4 vs salinity plots would help the discussion.
The whole discussion of methane in surface water has been re-written, and we now also include a figure with methane versus salinity. Line 373 ff

L346 "we thus exclude the Lena River as methane source" strong statement not based on quantitative analysis. In theory you should be able to calculate the CH4 input from rivers using concentration and discharge.
Ok, the wording was misleading, we did not want to make up a budget calculation. Thus we only conclude that the concept of salinity versus methane is not applicable for our data set.
L 380 ff

L347"One reason for this missing correlation, could be another source of freshwater, but with low methane concentrations." Formulated that way, it appears as a speculative statement.

L355 "This additional aspect of the water budget in ice covered estuaries might explain the missing relation between salinity and methane concentration." Ok, but please improve this part of the discussion (and associate figures) to make it clearer.
New L 382, the paragraph has been reworded

L456 "matter of debate between biogeochemists, ecologists, and PHYSICISTS" please revise English

Corrected

Comments to Referee 4

L8 : « biggest" in terms of what ? Discharge ? Drainage area ?
Changed to "largest river" concerning discharge

Please define the abbreviations (qPCR, MISA, etc…) in the abstract. The abstract should be understood on its own, which requires that abbreviations are defined. Refer to instructions "The abstract should be intelligible to the general reader without reference to the text", which is obviously not the case if the abstract is full of abbreviations. Since there is no size limit for abstracts in BG it should be not a problem to define all of the abbreviations.
The abbreviations are now replaced by the full name of the method.

L64: "MOBs" abbreviation is not defined, as it should the first time an abbreviation is used. Same applies to DOC, TDN, etc… The authors should take the time to polish the presentation of their work. It's not the reviewer's job to check this, especially at the second round of review.
MOB has been explained above in line 59. The DOC and TDN are now explained. In addition the Ms has been now polished by a professional editing service!

L74: Here, explain how the present paper adds to the previous papers by the authors in the area.
*In this study we focus more on the northern part of the Lena Delta and for the first time we were also able to measure the microbial methane consumption rate. New L76 "*The aim of the present study was to obtain an overview of the methane distribution in the northern parts of the Lena Delta and to gain the first key insights into the role of methane-oxidising bacteria (MOB) in the methane cycle occurring in this area"

L97: Specify what was the delay between the sampling and the analysis of CH4 in the home laboratory ? How were samples stored between sampling and analysis in the home lab: room temperature ? Did the authors check if poisoning with H2SO4 is efficient to stop biological activity and for how much time ? While I can imagine that acidification can preserve a sample for a few days, I not sure this is adequate for storage for months.
Sample preservation is always a critical point. However, our stoppers are rather methane save and a strong shift in pH is preferable to the more difficult to handle and environmental dangerous HgCl2. With a similar set up, Magen et al could show that methane concentrations in preserved samples with methane concentrations > 1 ppm did not change over a year [Magen, 2014 #2514]. Even though his shift in pH was obtained by NaOH. Samples poisoned with acid and stored in glass bottles with butyl stoppers also did not change in DIC content [Taipale, 2009 #2791].
We changed the text to:
The samples were stored upside down at temperatures < 15°C and analyzed after 4 months. Glass bottles and butyl stoppers are relatively methane tight and acidification of water samples results in good long-time sample preservation[Magen, 2014 #2514; Taipale, 2009 #2791]. However, we cannot exclude that some methane of the samples was lost.

L101: specify the concentration of the NaOH solution.
It was 1 M NaOH

L185: it's Liss not Lisa
Sorry, for the misspelling, in the references it was correct…

L 195: the relationship was not "developed for coastal seas", it was derived from an experiment in a continental shelf. In fact the Nightingale relationship converges with relationships derived in the Southern Ocean (Ho et al. 2006).
Ok, changed to "obtained for coastal seas"

L 235: "very high" is not adequate to describe CH4 concentrations since they vary by several orders of magnitude accross aquatic systems. 400 nmol/L might be "very high" for the Laptev Sea but will be very low for a tropical reservoir. Refer to actual numbers.
The actual numbers are now given, and changed to ". At station TIII-1304 we also observed comparably high methane concentrations in surface (212 nmol $L^{-1,}$, figure 3)

L 236: use an uniform unit throughout the text, a not a mix of µM, nM, nmol/L.
All concentrations are now given in nmol/L, only the fluxes are given in µmol/L/m2.

L 321: Did the vertical profiles of T and S show a mixing in response to "wind increase" ?
The water depth at this station was very shallow (4 m) and very "bumpy" at the time of sampling, thus no CTD cast is available, only information on the water of the Niskin bottle.
We also decided to remove the "outlier" TIII1304 from the discussion, but focus on the other stations. Section 4.1 Methane concentration and 4.3 diffusive methane flux have been restructured in a hopefully more coherent way.

L 327, 363: "Unfortunately" is a subjective evaluation. Scientific writing should neutral and objective.
Ok, has been removed.

L 333: Oxygen in surface waters of rivers/estuaries can also be lowered by sediment organic matter degradation, since sediment respiration is usually equivalent to water column respiration in estuaries (Gazeau et al. 2004). So the correlation of CH4 and O2 does not necessarily imply that CH4 is produced in the water column due to a process that consumes O2 in the water column.
Yes, but as we have a strong pycnocline separating surface and bottom water, and as O2 is only influencing CH4 concentrations in the surface water, we still think that in situ methane production is a possible methane source. See new Line 355 ff

L336: Is there evidence that DMSP occurs in the Lena river/estuaries ? The study of Florez-Leiva et al. 2013 shows that the increase of CH4 in response to a spike in DMS was 2 nmol/L/d. Could this production term sufficient to account for the CH4 concentrations ?
The process of in situ methane production is now discussed in more detail, but also stating that it is not clear yet, whether the experimental shown process really will result in elevated in situ methane concentrations.

L338-345: The CH4 concentrations in the Arctic estuaries (Lena, Ob and Yenisei) are much lower than those in temperate and tropical estuaries. This is an important information from this paper that should be mentioned and discussed. This probably results from the low temperatures and possibly lower organic matter concentrations in rivers (although this needs checking and discussing). This is relevant because temperature and possible organic matter concentrations in rivers can change in future in Arctic estuaries.
The low CH4 concentrations and CH4 emissions are now discussed in relation to temperate and tropical estuaries (L 506ff) and in the conclusion we extrapolate our data to future changes in the Arctic.

L 460: what is the reproducibility and accuracy of these measurements ? Is a 0.02 ppm difference significant, given differences in calibration gases, methods, etc… ?
Probably, this is no real difference, thus we state that our values within the range of literature values. L 481.

L 466: This not the case in the Southern part of the North Sea where there is no thermal stratification, and fluxes are much higher than in the Northern part of the North Sea where stratification occurs (Borges et al. 2016).
This references is now added to the text and the comparative table. Also the influence of stratification is discussed L504 and L516

L 520: diverse
corrected

Figure 2: remove PSU. Salinity is measured as a ratio of two conductivities (sample:standard), hence, unitless.
corrected

In the plot of Figure 2 replace nM by nmol L-1
corrected

In plot of Figure 3 replace nM by nmol L-1 and remove @ depth (m) = first
corrected

It could be useful to add the salinity plot (appendix figure) to figure 3, so that the CH4 concentration can be compared directly to the salinity (on the same figure).
We preferred to add a new figure 7, showing the methane concentration versus salinity, as suggested by the editor.

In plots of Figure 5 add the units of MOX, and remove @ depth (m) = first
corrected

Appendix Table A1: specify if this was done only for the surface data or the full data-set (all depths). Specify what do the bold numbers mean.
The table legend has been changed to:
Appendix Table A1. Linear correlation between the methane concentration versus different environmental parameters splitted into three water masses with their whole respective data set. Analysis was performed with log transformed data, shown are the r^2-values, the level of significance (p) and the positive or negative correlation (+/-), bold numbers indicate a significant correlation ($p<0.05$).

References
Borges AV, W Champenois, N Gypens, B Delille, J Harlay (2016) Massive marine methane emissions from near-shore shallow coastal areas, Scientific Reports, 6:27908, doi:10.1038/srep27908
Has been included now

Gazeau et al. (2004) The European coastal zone: characterization and first assessment of ecosystem metabolism, Estuarine, Coastal and Shelf Science 60, 673-694

Ho, D. T., C. S. Law, M. J. Smith, P. Schlosser, M. Harvey, and P. Hill (2006), Measurements of airsea gas exchange at high wind speeds in the Southern Ocean: Implications for global parameterizations, Geophys. Res. Lett., 33, L16611, doi:10.1029/2006GL026817.