

Anonymous Referee #1

I would like to thank the reviewers for their valuable comments, and the Ms has been restrutured and additional calculation were added. I believe that the changes made have improved the Ms substantially.

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General Comments: The data presented in the manuscript are important given the emerging role of inland waters and the global greenhouse gas balance. I think both the region of study and the data are important for understanding the response of northern ecosystems to climate change. I was surprised that many important papers regarding aquatic methane emissions were not included in this paper.

I was also surprised that no data regarding CO₂ concentrations were presented. In fact, CO₂ was not even mentioned; the third sentence of the abstract reads “With this melting, large amounts of carbon – either organic or as methane – will reach the waters of the Lena and the adjacent Buor Khaya Bay (Laptev Sea)”. Why would CO₂ not reach the Lena? This is a fatal omission by the authors as there is a rapidly expanding literature regarding CO₂ fluxes from northern aquatic ecosystems (some are just now going to press; i.e. doi:10.1029/2012GB004306, DOI: 10.1111/gcb.12083, doi: 10.1111/j.1365-2486.2009.02092.x, doi:10.1029/2008GB003404) Aside from an incomplete framing of the topic, my main issues with the paper concern the estimates of evasion, an incomplete description of the redox conditions and no attempt to constrain the proportion of meltwater (a proposed source of methane) to the river and estuary methane balance. If waters are oxygenated, how is methane produced in situ? Can meltwater balance the CH₄ in the river given the potentially enormous discharge of this river? I cannot support publication of this manuscript in its present form.

“The figures could be more clear. For example, Figure 1 is rather difficult to interpret. Coloring or shading of water and terrestrial portions would help, labeling of important features might also be useful. Where are: Muostakh Island, Buor Khaya Bay, Lena Channels, Olenekskaya Channel on the map?”

Two new figures (1a for the coast, 1b for the Lena Delta) have been prepared now which explain in more detail the locations.

Comments directed to the prescribed questions: 1. Does the paper address relevant scientific questions within the scope of BG? Yes, I find that the paper addresses a relevant topic that is likely of interest to readers of BG. 2. Does the paper present novel concepts, ideas, tools, or data? I believe that the data is important. 3. Are substantial conclusions reached? I do not find that the conclusions are fully supported, and are thus not substantial in present form. 4. Are the scientific methods and assumptions valid and clearly outlined? I do not find that all of the methods are supported. Assumptions regarding water sources (especially the meltwater balance) are not supported in the text. 5. Are the results sufficient to support the interpretations and conclusions? I do not find the results to be sufficient. The interpretation of the mixing experiments is especially problematic. 6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? I find the methodology to be sound. 7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution? The authors properly credit other works, however, there are significant omissions of relevant works regarding aquatic methane and CO₂ fluxes. 8. Does the title clearly reflect the contents of the paper? Yes 9. Does the abstract provide a concise and complete summary? I do not find that the abstract is supported by the data and the analysis described in the text. See detailed comments below. 10. Is the overall presentation well structured and clear? I do not think the organization of the paper is strong. The introduction requires more background and references to the literature. Certain components of the methods section are not placed properly (i.e. the study site), and the section of the discussion regarding gas transfer velocities actually belongs in the methods section. 11. Is the language fluent and precise? The language is mostly precise, however, I would suggest a final review by a fluent speaker before resubmission. 12. Are mathematical formulae, symbols,

abbreviations, and units correctly defined and used? 13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? I think the figures could be reformulated. Figure 1 is specifically problematic (comments are provided above). 14. Are the number and quality of references appropriate? No, there are serious omissions of references regarding aquatic gas fluxes in northern regions. More background is especially required in the introduction. 15. Is the amount and quality of supplementary material appropriate? NA.

Detailed Comments:

16214.2: My understanding is that the permafrost community prefers the term “permafrost thaw” as opposed to “melt.” The term melting implies water ice. I would recommend that melt be replaced with the thaw term. Alternatively, a description or discussion of ice content in the region is necessary to support the hypothesized meltwater methane source.

The term “melting” has now been replaced by “thawing” and the location of water sampling at the permafrost cliffs has been termed “creeks draining from permafrost soil”.

16214. 5: Will any of this carbon reach the Lena as CO₂?

Yes, of course does CO₂ also reaches the Lena and the Laptev Sea. This is reported by Semiletov et al 2011, 2013. The intention of this study was to focus on the methane aspect. However, the aspects of the CO₂ input have now been included into the introduction and abstract.

16214. 16: Where is the methane released? In the bay or in the river itself?

This statement refers to both sites. This statement has been added to the abstract.

16214. 19: Unclear. Suggestion “10% of global runoff flows into areas of shallow shelf seas.”

Has been changed accordingly

16215.2: I understand that runoff may change, but specify the likelihood of increase, and the expected magnitude.

Some more detailed information on the increase of river discharge, its reason and its implications have been added.

16215.11: transition between paragraphs. Perhaps begin by stating the relevance of CH₄ in regional budgets and the large uncertainty.

This paragraph has been rewritten.

16216. 16: Sampling details should be included in the following section

A new section for the study site was added here. For the procedure of water sampling however, I added some more details, however I think that all main information is already given.

16216.16: what gas was used to flush the serum bottles?

The bottles were flushed with sample water to assure that the “final” water in the bottle has not been in contact with the atmosphere to avoid gas exchange.

16218.4: Please explain the purpose of these mixing experiments initially. I am concerned that these experiments to determine aerobic methane oxidation actually experienced anoxic conditions. The oxygen environment must clearly be explained. If not, these results can not be interpreted as the authors present them. The issue of oxygen conditions and methanogenic potential is currently a major problem for the conclusions of this manuscript.

I agree, that a methane oxidation under oxic conditions is difficult to explain, even though not impossible. Thus I decided to delete the experiments from the manuscript. It is now only stated that simple incubation experiments did not reveal any methane oxidation.

16219.3: what were the oxygen conditions of the sampling locations? The methane production potential (3.4 Mixing Experiments) is essentially irrelevant if the redox state will not support

methane production.

The in situ oxygen concentrations of the sampling location are now given in the text and the experiments are deleted.

16219.17: remove “in a first overview”. What area are we talking about? The River, the estuary? State it clearly. I think it would be useful to separate Figure 3 into river and estuary samples. Probably it is better to show the data on an “aerial” basis and split them as suggested.

A new figure 3 has been prepared now, where the data are split into creek meltwater, river and estuary, as well as the different years. For the estuary new figure 4 have been made where the stations and the corresponding data are plotted within a “common area”. The text has been changed accordingly.

16219. 23: Is there any explanation for the higher methane concentrations ($\approx 100\text{nM}$) at greater depths shown in the lower panel of Figure 4?

The blue data points in the lower panel refer to station within the Bykowski Channel which is rather deep, but the corresponding surface values are within the same range. I also tested all data sets, but there never was a statistically significant difference between surface and bottom data.

16219.25: The repeated reference to the “background” concentration of 20nM needs to be evaluated further. Is this concentration reflective of waters in equilibrium with respect to the atmosphere or is the water supersaturated/undersaturated?

The paragraph on the methane concentration has been rewritten and the word “background” is now omitted, instead the median of the respective areas and years is given.

The waters were always oversaturated with methane, around 900%. These data are given now in the text.

16220.20: Omission of these data is surprising given the stated purpose of the paper. Could non-biogenic sources such as those referred to in the introduction be important? What processes could account for these signatures? Degassing, hydrates, something else?

The observed isotopic data are now discussed more detailed, including the occurrence of the heavy “outlier”. Cramer and Franke report also a rather heavy signature, and report fracture zone where thermogenic methane could escape.

16221.15: what sources do these ^{13}C signatures reflect? I appreciate the comparison to literature values, but more complete interpretation is necessary.

A more detailed comparison of the isotopic data with literature is now added, see page## and ##

16222.3: I liked seeing the downstream methane pattern in Figure 7. This result is rather interesting. This result could be extended by modeling gas residence time/distance. For example given the concentrations at 0km , what would the predicted concentration be at 2km downstream, or at further distances. The authors suggest that there are new sources but this statement must be proven more explicitly. This analysis should be extended further to the estuary data, assuming advective flows into the estuary, how far out would you expect to find a riverine methane signal before all the methane has been lost to the atmosphere? I suggest the equations reported by Baulch et al (2011) (and references therein) for 95% evasion length (ie. $3v/k$), or a simple model of exponential decay would be a good starting point.

I included now a simple box model and tried to estimate the methane input through the creeks. To estimate the input of the riverine methane into the estuary the equation of Bauch et al would certainly be useful, however data on the current regime or flow velocity are crucial for this equation; and these data are not available. Thus I estimated the riverine input and compared in to the diffusive flux into the atmosphere.

16222.12: Discussion of methane emission modeling belongs in the methods section. Furthermore, given that evasion is the main process described in this paper, a more detailed discussion and evaluation of gas transfer is required. It is unclear why this particular model was chosen, or if it is even applicable in this environment. The physics of gas transfer are perhaps the most uncertain components of evasion estimates, and thus require much, careful attention. How do the estimates of k

compare to other literature values?

The k values have now been recalculated according to more modern literature. Also the k600 value is given in now in comparison with other literature values.

16222.20: from where does this transfer velocity equation originate and why this particular equation? There is a large literature regarding transfer velocity modeling in rivers and it would be important to evaluate other potential equations and their application to the studied waterbody.

The description on the calculation of the methane flux including the transfer velocity is now moved to the method section, and in the discussion I added a comparison of different k600 values.

16223.6: This statement is important. The comparison between terrestrial and aquatic emissions is important for understanding the landscape greenhouse gas balance and for predicting future change. However, I do not believe that the data can support this conclusion. Seasonal variability could alter this conclusion and I do not believe that these data can support a direct comparison between the two ecosystems. How much variability was there in the eddy covariance data cited?

I did not intend to directly compare the different ecosystems, because then the aerial extension of each system would have been taken into account (as is now stated in the text and for which no data are available to my knowledge). Most studies in the Arctic have been done in summer (at least the cited studies), thus the seasonal aspect is only minor. The comparison of river channels with ponds and terrestrial environment was only intended to show the magnitudes of other fluxes.

The range of the eddy covariance data is now added.

16223.14: The statement “we can only conclude that the observed methane concentrations of around 100 nM are the result of a strong meltwater input and a strong in situ production of Methane” is highly problematic and is not well supported. Most importantly, the redox status of the sampled waters are not presented. If oxygen is present, in situ methane production would not be probable. The suggested methane sources are not fully supported. Please include estimates of discharge from the Lena and provide at least some discussion of water sources. The meltwater source is not supported hydrologically in the text. Given the expected large flows in the river would there be enough meltwater input to detect new CH₄ in the river? The author does not present any data regarding river discharge and it is difficult to judge whether small inputs could even be detected. Could groundwater or production in the river sediments be an important methane source?

The conclusion of the river section is now transferred to the final conclusion.

The experiments on the methane cycle are now omitted and I only state that there are indication for in situ methane production. Hydrological evidence of the input of meltwater is now given.

16224.13: What might explain the outliers plotted in Figure 9?

“Cramer and Fisher also report methane escaping from a SW - NE fracture zone in the Laptev Sea. This could also explain the very heavy signature (-10‰) of the three outliers (Fig. 5) as gas having escaped by the fracture zone.”

16224.21: Is there any literature that describes controls on methane oxidation in other estuaries?

No, not much work is done regarding methane oxidation in estuaries, except the early work of de Angelis.

16225.10: Again, reference to high methane production are not supported by oxygen data. I assume that the river and the estuary are oxic, so how can methane be formed?

Eventhough methane production is a anoxic process, there are several possibilities and references that it also can be produced in the presence of oxygen, either by other precursors (DMSP) or anoxic microniches. However, this discussion would lead to far, thus the statements on methane production are reduced, and we hope to make up new experiments to prove the indications.

16225.15: The author does not adequately present conclusions regarding the mass of methane which could reach the bay. What fraction of the methane is emitted before reaching the estuary?

An additional mass balance of riverine methane input into the bay versus diffusional loss is now included.