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Biogeosciences Discussions

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10, C8854–C8870, 2014

Interactive Comment

Interactive comment on "Pumping methane out of aquatic sediments – forcing mechanisms that affect the temporal dynamics of ebullition" *by* A. Maeck et al.

A. Maeck et al.

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Comments of the anonymous referees. Authors responses were marked by "Autors response:"

Anonymous Referee #1 Received and published: 16 January 2014 General comments This study investigates controls and mechanisms of CH4 ebullition from a heavily human-impacted river in Germany. The authors have made high-resolution measurements and use these to investigate the temporal variability of bubbling events and how these are linked to atmospheric and hydrostatic pressure changes. Overall, this is a good piece of science that is relevant to the scope of BG. The study does not present

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novel concepts, but its technical aspect and analysis of data make it an important contribution to the understanding of the complexities of ebullition. The manuscript is well structured, the language is fluent, and the choices of references are appropriate. There are, however, studies published in 2013 that also use large datasets to investigate variability in CH4 ebullition from freshwaters. The scientific method and the aim of the study are outlined in the introduction and followed up throughout the paper. Calculations and mathematical formulas are properly described in the method section. The methods are to some extent outlined clearly, but they need revisions when it comes to the analysis of CH4 and to the approach when determining average CH4 concentrations in the bubbles (see specific comments). The main concern regarding the content is the extrapolation of measured CH4 emission (in the very end of the manuscript) which should be removed entirely in a revision. The authors suggest that, based on their results, the global estimate of CH4 emissions from freshwaters could potentially be underestimated by 50%. This may very well be true, however, such a suggestion should not be made based on the results in this study. These measurements were made in a system that is heavily influenced by human activity and therefore not at all representative for freshwaters globally. The authors point this out in the same section, a statement that is contradictive and does not support the extrapolation of measured emissions.

Specific comments Title: The title is too general. This study is looking at a river that is heavily influenced by human activity. It is not representative for any ("aquatic sediments") natural system.

Authors response: To the title, the part “in an impounded river” was added.

Pg 18688, ln 9: Shouldn’t “mechanisms” read “trigger”?

Authors response: Yes, this was changed.

Pg 18688, Ln 15: Why only “underestimate”? Short sampling intervals are also likely to overestimate. Also, the extrapolation should be taken out of the abstract and out of the manuscript (see comment below).

Authors response: The global extrapolation was taken completely out of the manuscript. We wrote “likely underestimate” because the chance to underestimate the flux is higher than to overestimate the flux (Fig. 6). This is because rare, but large ebullition events have a significant effect on the average flux rate.

Pg 18688, Ln 22: State a range in average bubble CH4 concentration. There are many papers that report different values and there are papers that report large spreads, e.g. Wik et al. 2013, JGR-Biogeosciences. a Pg 18689, Ln 21: The fate of rising bubbles is to some extent understood during the ice free period and in shallow regions, but certainly not in deep zones and during winter when lakes and even many rivers are ice-covered.

Authors response: Yes, in these cases, the transport is more difficult. For the deep zones, validated bubble dissolution models exist (e.g. McGinnis 2006, Leifer 2003) which can help to predict the fraction of the bubbles CH4 dissolved during its rise. However, in impounded rivers, the water depth is mainly shallow (<10m, most often $\sim$ 4m) therefore, the bubble dissolution is limited. For large and deep reservoirs, this can change dramatically, especially in case of thermal stratification. Regarding the fate of bubbles during the ice-covered period, we cannot say anything about this because there was no ice cover during the study period. In the River Saar, ice-coverage is seldom and occurs only partially in some years.

Pg 18690, Ln 1: There are additional papers that may be cited here that report large datasets on CH4 ebullition from freshwaters, e.g. Wik et al. 2013, JGRBiogeosciences.

Authors response: Thanks for that comment. We included the reference.

Pg 18691, Ln 24: Why were all measurements made in areas where you knew there would be high fluxes? It is as important to investigate variabilities and forcing mechanisms in zones where high fluxes are not expected, especially when aiming to extrapolate results. The river Saar is most likely not representative globally. Hence, results should not be extrapolated as if measurements were made in an undisturbed natural system (see comment below).

Authors response: The deployment sites were chosen, so that different flux magnitudes could be covered. Unfortunately, it was not possible to deploy an ABT at a site with an even lower expected flux rate since it would be the main cargo ship channel. And yes, the River Saar is not representative for all aquatic systems and therefore, we excluded the extrapolation. However, there is still a lack of data from other system types, for example from tropical impounded rivers. It would be very interesting to compare more impounded rivers with each other and also with results from other systems which use continuous ebullition measurement methods.

Pg 18693, Ln 6: Why do you base all your flux calculations on CH4 concentrations measured in bubbles that were deliberately stirred up from the sediment and not spontaneously released?

Authors response: Spontaneously released bubbles must rise through the entire water column and are in exchange with the ambient water. Additionally, to collect the spontaneously released bubbles, a bubble trap must be deployed for a longer time span which favours a gas exchange within the gas capturing unit. To reduce these errors, we connected a weight to an funnel and caught the released bubbles immediately. So, dissolution effects could be minimized.

Pg 18694, Ln 3: This refers back to the previous comment. An average bubble CH4 concentration of 80% is a lot. Again, bubble CH4 concentrations often vary greatly in both time and space.

Authors response: Yes, we know that it is often reported. However, in the River Saar, we have a system, where large amounts of organic carbon is stored in the sediments under anoxic conditions (oxygen concentration in less than 1mm depth below the sediment-water interface is 0%), nitrate, manganese, iron and sulfate is also very limited and only measured in the upper 3 cm of sediment cores. Therefore, the largest depth interval of the sediment column favors methanogenesis. Of all available gases within the sediment, CH4 has the lowest solubility and is therefore the first gas which exsolved if the partial pressure of all dissolved gases exceeds the ambient pressure. Therefore, we think that most bubbles leaving the sediment have very high CH4 concentrations and that the difference between 100% and the measured concentrations are mainly caused by bubble dissolution. In other systems, e.g. lakes, other processes like O2 production in macrophytes or N2 generation by intensive denitrification can also produce free gas. However, in the Saar, no macrophytes are present and denitrification rates are supposed to be much lower than CH4 production rates and therefore, these processes may play only an insignificant role.

Pg 18694, Ln 21: Did you compare the concentrations in bubbles from disturbed sediment to the concentrations in spontaneously released ones? This is important when using stirred bubble concentrations in flux calculations and using an average value as high as 80%. Also, how often were the sediments disturbed for bubble CH4 concentrations?

Authors response: No, we did not compare it in this study. But former measurements showed that dissolution during the deployment of normal funnel shaped gas traps is significant. Therefore, we decided to used deliberately released bubbles for the concentration measurements (see also comment above). And I agree, that the CH4 concentration is important in terms of the overall flux. However, most variability is in our study based on the variability in the released volume. Hence, the concentration is of minor importance regarding the trigger mechanisms of ebullition.

Pg 18695, Ln 4: More information on the analysis of CH4 would be useful. GC method specifications (e.g. temperatures and flows) should be made explicit.

Authors response: We added more details about the GC method and used components (Material and Methods).

Pg 18697, Ln 5: The measurement period should also (or instead) be made explicit in the methods.

Authors response: The measurement of discharge were carried out by the Federal Institute for Hydrology, BfG, Koblenz, Germany. We included that the values refer to the average daily discharge rates.

Pg 18698, Ln 18: Are the fluxes normally distributed? If not, consider using percentile ranges as a measure of variability.

Authors response: Thanks for this comment. The fluxes are not normally distributed, so we changed the mean to the median and reported 25 and 75-percentiles instead of the standard deviation.

Pg 18698, Ln 26: Where was the mooring located in relation of the trap? State that in the methods.

Authors response: The anchor weights were placed 7-9 m away from the ABTs and set so that the ABTs were always at the same location. We added a more detailed description to the manuscript.

Pg 18701, Ln 25: Production of CH_4 is important. See comment below.

Pg 18702, Ln 23: This also implies that the recharge of gas in the sediment is an important control on temporal variability. Hence, sediment temperature and CH_4 production rates do play large roles. Low production between large events affects both emission frequency and amplitude.

Authors response: We fully agree, that the CH_4 production rate, which is also controlled by temperature, plays a role by determining the time between ebullition events. If not enough gas can be "refilled" by CH_4 production, the next forcing may not release bubbles. And the longer the period between two forcing events and the higher the CH_4 production rate, the larger will the ebullitive flux be during the forcing event. This issue is discussed in the discussion section (4.2) of the manuscript.

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Pg 18704, Ln 13: What is the difference in flux magnitude between day and night (i.e. in this case human induced pressure changes vs. more natural)?

Authors response: The difference between day- and nighttime flux rates is large and there is a significant difference. We included a table showing this (with statistics) and added a paragraph in the methods section (analysis) and results section.

Pg 18704, Ln 25: The effect should vary whether it is a shallow or deep lake and with bottom topography.

Authors response: Yes, in shallow regions of a lake, the proportion of the pressure change is much stronger compared to deeper zones. Therefore, the same wave will probably have a higher potential for releasing bubbles in shallow water compared to deeper parts of the lake. Studies, e.g. Bastviken 2004, showed, that shallow areas emit more CH_4 via ebullition compared to deeper zones and therefore, the trigger of ebullition via waves or seiches is probably more important for shallow areas.

Pg 18705, Ln 1: Shouldn't "control" read "trigger". The production rate in the sediment acts as the ultimate control of ebullition.

Authors response: "Control" was changed into "trigger".

Pg 18707, Ln 7: The global extrapolation should be removed from the manuscript. The study is made in three locations only and in a system that is not representative for aquatic systems globally.

Authors response: The extrapolation was completely removed from the manuscript.

Table 1: Why is the average concentration (48.6%) in January at ABT-1 much lower than the rest?

Authors response: We do not have an explanation for this. Possible causes could be that there was an error during the sampling. Since all replicates show no divergence, a possible error could have only been made during the gas collection. Possible sampling error can only reduce the CH4 concentration, since the dilution with ambient air leads to this pattern. However, because one value is an outlier, we did not remove it completely from the results.

Fig. 2.: Why not have volume on the x-axis if it is a “volume determination” error?

Authors response: We decided to report the error depending on the ebullition volume, because it is for interpretation of the results much more helpful, to report the error as shown. Because then, the results can be immediately checked for accuracy.

Technical corrections Pg 18689, Ln 9: “methane” should read “CH4” for consistency

Authors response: corrected.

Pg 18689, Ln 12: “methane” should read “CH4”

Authors response: corrected.

Pg 18691, Ln 19: “methane” should read “CH4”

Authors response: corrected.

Pg 18695, Ln 8: Specify “five months”

Authors response: corrected.

Pg 18701, Ln 24: “mechanical forcing” should read “anthropogenic mechanical forcing”

Authors response: corrected.

Pg 18702, Ln 8: “methane” should read “CH4” Authors response: corrected.

Anonymous Referee #2 Received and published: 17 January 2014 General comments: This paper addresses the temporal character and triggers of ebullition from sediments in an impounded river, relevant questions within the scope of BG. The authors collected methane ebullition data using 3 automated bubble traps with frequent sampling and over 5 months in regions of known high sediment accumulation. The paper provides novel quantitative evidence for the role of self-inhibition in ebullition, perhaps due to depletion of the trapped bubble store in the mud. In addition, the authors propose a novel framework for comparing the temporal character of ebullition from different settings. The analysis of how uncertainty in flux estimates grows with shorter measurement periods is clear and illuminating, but the extrapolation to global emission estimates is unwarranted due to the small sample and its bias. The writing is clear and fluent, but some of the text was inconsistent with the data presented in Table 2. The references are appropriate, and the abstract is concise and complete.

Specific comments:

Title: "Aquatic sediments" should be replaced with something more specific to the study site.

Authors response: To the title, the part "in an impounded river" was added.

Pg 18694, Ln 4: Have you done anything to estimate loss rates from dissolution of gas trapped in the ABTs? Since you claim to be able to measure ebullition rates within 10%, you should check that dissolution does not significantly increase that number. It would then be appropriate to note how this uncertainty propagates to your flux estimates and conclusions.

Authors response: Yes, we checked for dissolution theoretically using Fick′s Law. With a CH4 concentration of 80% in the gas phase, 1.8 μM in the water column and a piston velocity of 1 cm h-1, the dissolution removes within one hour 1.8*10-4 ml, which is much less than one μl and therefore does not affect the accuracy of the sampling. However, since bubble can cause strong turbulence, we checked it again with a piston velocity of 100 cm h-1, which leads to a volume error of 1.7*10-2 ml, which is still insignificant.

Pg 18694, Ln 20: The gas disturbed from the bottom could have a different chemical composition from bubbles released naturally.

Authors response: For determining the CH4 concentration of the bubbles, there are

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two possible ways, either (1) deploying a gas trap for longer timespans (hours) or (2) releasing bubbles deliberately with catching them immediately. We've chosen method (2) since with method (1), the bubbles must rise through the entire water column and are in exchange with the ambient water. And since the bubble trap must be deployed for a longer time span which favours a gas exchange within the gas capturing unit, the error increases. To reduce these errors, we released the bubbles deliberately and caught them immediately. So, dissolution effects during the rise and during the deployment time could be minimized. In addition, the focus of this manuscript to study the variability of ebullition. And since this is mainly determined by the variability in released gas volume, possible errors arising from concentration uncertainties are of minor importance.

Pg 18696, Ln 14: The choice to do logical regression seems interesting. The rationalization that the volume of gas released is not a linear function of the forcing mechanism suggests that you performed a regression analysis on the gas flux data themselves. What were those results? It would be interesting to see which forcings correlate well with the actual fluxes, taking into account their widely-varying magnitudes. For example, what if high-frequency hydrostatic oscillations allow low-level ebullition (still higher than the mean over the measurement period), while low-pass hydrostatic pressure drops trigger large venting events?

Authors response: We tested several approaches before we decided to use the logistic regression. But due to the non-linearity and complexity of ebullition, the logistic regression approach simplified the response variable. However, since the mechanism seems to be still too complex to analyze it quantitatively and it yields no additional findings, we removed the logistic regression analysis completely and added the comparison between day- and nighttime flux rates.

Pg 18699, Ln 19: Analysis of synchronized 5-min ebullition events assumes that the forcings also trigger the ebullition within the same 5-minute period. This may be true, but some experiments in mud-analog systems have shown that mobile bubbles may

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take a few minutes to rise to the surface (Boudreau et al. 2005). What happens to the percentages and regression coefficients when you allow for time lags of 5, 10, or 15 minutes?

Authors response: That is right. Therefore we did not use the 5-min period for the analysis but the average over one hour. So, these effects are included.

Pg 18700, Ln 3-5: The logistic regression can identify impacts on the presence or absence of significant ebullition, but that's not as general as explaining the "observed variability of ebullition rates."

Authors response: The logistic regression analysis was completely removed.

Pg 18700, Ln 5-6: The text suggests that high-frequency hydrostatic pressure variations decrease the probability of high ebullition rates. However, Table 2 shows that 2 of 3 traps gave positive regression coefficients with high frequency pressure fluctuations. The remainder of the quoted paragraph similarly supports a positive correlation between high-frequency fluctuations and higher ebullition rates, though later text contradicts this (see comment on pg 18702 below). Authors response: The logistic regression analysis was completely removed.

Pg 18700, Ln 19-21: You observed that the site with fastest sedimentation had the highest mean ebullition rate. What were the mean ebullition rates for the 3 traps, and how did they compare with the exponential relationship found in Maeck et al. (2013)?

Authors response: We included the results of the exponential fit in the discussion section.

Pg 18702, Ln 17: This discussion again references negative regression coefficients with high frequency pressure fluctuations and provides a plausible explanation for such results, but the text contradicts Table 2.

Authors response: The logistic regression analysis was completely removed.

Pg 18703, Ln 4-25: What were the water depths where the 3 ABTs were deployed? If shallow sites can generally release bubbles in response to ship wakes, but ABTs 2 and 3 did not vent anything following ship passages, it's important to your explanation to know if 2 and 3 were deeper than 1. I couldn't find those numbers in section 2.2 or Fig 1.

Authors response: We added the water depth of the sampling sites in the study-site description and included in the discussion the possible effect of the water depth.

Pg 18705, Ln 1: Varadharajan and Hemond (2012) showed that ebullition was triggered by drops in hydrostatic pressure, not just atmospheric.

Authors response: "hydrostatic" was added.

Pg 18705, Line 9-29 and Fig 9: The proposed framework to compare and understand regimes of episodicity in ebullition is a valuable contribution of this study, but I wonder if it could be clarified and simplified. The essential distinction supported by the data is between regimes of external triggering vs. fast internal methanogenesis that makes bubbles large enough to be mobile faster than the trigger can release them. If you were to scale the time axis in the subfigures by the inverse forcing frequency, might not subfigures (1) and (4) have a similar temporal character? Both seem to be driven by external triggers, rather than an internal methane source. In the case of data from this study (1), the production rate and forcing frequency are both higher than in (4), but in both, one would need to sample over a time period much greater than the period between significant trigger events. These two cases seem similar to each other but distinct from (2), which you suggest is driven by its internal production rate because the forcing does not act frequently enough to relieve the buildup of gas bubble volume and buoyancy. While the conceptual example drawn in subfigure (3) seems plausible, it does not constitute evidence supporting this framework. Even if it were correct, I hypothesize that if one scaled the time axis by the inverse forcing frequency and magnified the y-axis to see temporal variability, the temporal character would be similar to that in (1) and (4). This is because such a case would also be externally-triggered, with the frequency of ebullition determined by the forcing frequency and the magnitude scaled with the methane production rate. To distinguish between cases (1,3,4) and (2), the externally-triggered vs. internally-driven regimes, one could simply consider the ratio of timescales of production vs. forcing. When the ratio of (production rate / forcing frequency) is low, one might expect an externally-triggered system, while a high value of that ratio would imply an internally-driven system.

Authors response: Thank you very much for these suggestions. It is right, that if the ratio between production rate and forcing frequency is used then all necessary variables are included and it is more simplified. Therefore, we changed Fig. 9 so that it is now one-dimensional and changed the text describing the conceptual framework.

Pg 18707, Ln 6: Given that your heavily-impacted study site is "not representative for all aquatic systems," it is unreasonable to extrapolate your results to the global scale.

Authors response: We removed the global extrapolation.

Table 2: Are the signs on the coefficients for High frequency pressure fluctuations correct? They do not match the text (see notes above). Authors response: Tab 2 was removed.

Fig 2: How does the uncertainty analysis shown in Fig. 2 impact the general results and conclusions? The asymptotic 4% relative error seems well-constrained, but how does that uncertainty propagate into flux estimates? One interpretation is that the uncertainty associated with short sampling intervals is more important than that from the instrument, but I don't see this stated in the manuscript. If that's the case, do you need a figure devoted to instrumental uncertainty?

Authors response: Within this study, the focus is on the mechanisms controlling the temporal variability of ebullition. Hence, the accuracy of the magnitude of the ebullition events is not as important as in studies, which assess the GHG emissions of an aquatic system. However, for all flux estimates where the magnitude is important we used the hourly and daily flux rates, which are much more accurate and hence, the instrument uncertainty is strongly reduced.

Fig. 9: Are the y-axes in the subplots scaled the same, by the methane generation rate, or something else? See note above.

Authors response: The Y-axes refer to the same scale.

Technical corrections: Fig 4: Units in subfig (a) should be mmol2 m-4 d-2 Hz-1 = mmol2 m-4 d-2 s, and in (b), Pa2 Hz-1 = Pa2 s

Authors response: Changed.

Fig. 8 could use more distinct line styles to distinguish between the ebullition from different traps (maybe consistent with fig. 5?)

Authors response: Since there is one additional parameter displayed, an additional color is necessary. We decided to change the colors for a better visualization. Anonymous Referee #3 Received and published: 22 January 2014 The manuscript from A. Mack et al on the "pumping of methane out of aquatic sediment“ is well written and concise. However, as it is submitted to a biogeochemical journal I do have some comments. There is a strong emphasis on physical parameters and their analysis, however the biogeochemical background is somewhat neglected. The analysis of their measured data is very extensive, but the interpretation of the data in an environmental context is a bit weak, and should be improved. Also, as to my knowledge, the described way of measuring the ebullition rate is new; a bit more details to the sensors and their calibration should be given. Especially when the huge amount of data is analyzed very extensively. I also found the proposed scheme in figure 9 a bit farfetched. The data and interpretation given in this ms, do not lead to this scheme. More general and specific comments are given in the modified pdf-file.

Authors response: Thank you for the comment. Since all other reviewers stated, that the content of this manuscript fits into the scope of BG and since the physical dimension is important for measuring representative fluxes, we think that the content is appropriate for BG. The way of measuring the ebullition rate is not new and unpublished. Varadharajan et al. (2011) described the principle in detail. The conceptual framework was stated as a “valuable contribution” by the other reviewers. But following their suggestions, we revised the Fig. 9 as well as the text. Thus, we present only real measured data in that scheme and it is now more clarified. Response to the commented pdf-file:

Authors response: P 18690, l28: The “review the methodologies” part was removed, since the manuscript describes the limitations of discrete sampling in contrast to continuous sampling and therefore not different instruments.

Authors response: P 18692, l1: The sedimentation rate is given later in the manuscript. In the referred part, the numbers refer to the sediment accumulation which gives the reader a more intuitive impression of the sediment thickness at the sampling sites.

Authors response: P 18693, l2: The fill height is described at the same page in line 14-15.

Authors response: P 18693, l9: We denote the ebullition rate in mol since it is then pressure independent and the direct relation to the emission of GHG is given.

Authors response: P 18693, l13: 1) The calibration refers to the pressure on the sensor which is directly correlated with the volume of gas within the sampling container. Continous bubble streams do infer the measurement only minorly, since the rising bubbles release their content in the container and therefore no turbulence of water is in contact with the sensor. 2) The drift of the sensor is given by the manufacturer of < 0.5% per year. 3) All ABTs were deployed in the same depth from the water table but over different water depths. The pressure and therefore volume change of gas bubbles was taken into account (see comment above). 4) We had no problems with sedimentation since all open parts were directed downwards. There was biofouling on the funnels itself, but the gas system (most of the time filled with a high content of methane) remained clean.

Authors response: P18696, l 13: We removed the analysis with the logistical regression completely.

Authors response: P 18701, l5-7: Thanks for this suggestion. We included the values.

Authors response: P 18701, l13: This is shown in Fig. 4, 5 and 8.

Authors response: P 18701, l23: We included the proposed addition.

Authors response: P 18701, l25: This point refers to "short" timescales. Hence we added "on timescales of days to weeks" to this sentence.

Authors response: P 18702, l7-8: The water temperature and its temporal pattern regulate strongly the sediment temperature. During periods of cold water, the sediment temperature increases with depth, while during the summer, the pattern is vice-versa. See for example Fang and Stefan 1996 or 1998.

Authors response: P 18702, l13: The analysis based on the logistic regression was completely removed and hence, the discussion of the gas content of the sediment changed.

Authors response: P 18702, l28: The section was removed.

Authors response: P 18703, l21: This information was added in the study site description.

Authors response: P 18705, l8: Pressure changes can be induced anthropogenically but also naturally. However, the result remains the same and therefore, the differentiation of the pressure changes should be made on the frequency and not the cause.

Authors response: P 18715: We included the march gas sampling since ebullition data ranged up to march 6. Additionally, one sampling point more makes the results more robust.

Authors response: P 18716: Relative pressure means that it the pressure difference to a reference pressure. We used this because then the results depend not on the water depth of the deployed pressure sensor and both pressures are comparable.

Interactive comment on Biogeosciences Discuss., 10, 18687, 2013.