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Biogeosciences
Discussions

Interactive comment on "Organic matter and sediment properties determine in-lake variability of sediment CO_2 and CH_4 production and emissions of a small and shallow lake" *by* Leandra Stephanie Emilia Praetzel et al.

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Authors' response to referee's comments (RC2) on the manuscript, "Organic matter and sediment properties determine in-lake variability of sediment CO2 and CH4 production and emissions of a small and shallow lake" by L.S.E. Praetzel et al.

Dear reviewer, Thank you very much for your comprehensive comments on our manuscript "Organic matter and sediment properties determine in-lake variability of sediment CO2 and CH4 production and emissions of a small and shallow lake". We

appreciate your suggestions both in form and content and believe that they will substantially improve the paper. In the following, we will outline our responses to your comments one by one and thereby hope to clarify open questions and incorporate your suggestions to your satisfaction. Each answer will be structured as follows: (1) comments from referee, (2) author's response, (3) author's changes in manuscript. Kind regards, Leandra Praetzel & Co-Authors

General (1) The manuscript needs careful line editing to take care of non-idiomatic English. An example is the frequent usage of wrong tenses (e.g. in line 60: "is mainly depending on" rather than "mainly depends on"). The authors may seek help from a native English speaker for this purpose. I have pointed out a few instances below, but these are by no means exhaustive. I must also concede that I am not a native English speaker! (2) We asked a native speaker for help and feel certain that her corrections will substantially improve the grammatical style of the paper.

Specific (1) Lines 14-15: Change "... to the atmosphere, following recent studies this is particularly the case for small and shallow lakes." to "... to the atmosphere; recent studies have shown that this is particularly the case for small and shallow lakes." (2) The sentence will be rephrased in the revised manuscript as follows: (3) "Inland waters, particularly small and shallow lakes, are significant sources of carbon dioxide (CO2) and methane (CH4) to the atmosphere."

(1) Line 16: Delete "yet" and "thus". (2) The terms will be deleted in the revised manuscript.

(1) Lines 21-22: Change "... were significantly negative ($p<0.05$, $rho<-0.6$) correlated" to "... exhibited significant negative correlation ($p<0.05$, $rho<-0.6$)". Please make similar changes elsewhere. (2) The sentence will be rephrased according to the reviewer's suggestion. Similar changes will be made elsewhere in the revised manuscript.

(1) Lines 32-34: The last sentence states the obvious. Who has suggested such a "replacement"? (2) Some studies implicitly equal production and emission

rates, e.g. Grasset et al. 2018: doi: 10.1002/lno.10786 Sollberger et al. 2014: https://doi.org/10.1007/s00027-013-0319-2 We will adjust the statement in the revised manuscript as follows: (3) “We highlight that studies on production rates and sediment quality need to be interpreted with care in terms of deducing emission rates and patterns as it this neglects physical sediment properties and production and oxidation processes in the water column.”

(1) Line 52: Change "has been" to "have been" (here majority is plural), and remove "is". (2) The term will be changed in the revised manuscript and “is” will be deleted.

(1) Line 56: Remove hyphen between "in" and "lake". (2) The hyphen will be removed in the revised manuscript.

(1) Lines 58-59: Why is it crucial? Your results show that it is not. (2) The sentences will be rephrased in the revised manuscript as follows: (3) “Nevertheless, anoxic sediments are important for whole lake C cycling as the CO2 and CH4 produced there can be released through the water column to the atmosphere. To understand the spatial patterns of CO2 and CH4 emissions, it is therefore of interest to also understand CO2 and CH4 production processes in the sediment as well as their major controls.”

(1) Line 64: Also its origin (e.g. lignin). (2) This will be added in the revised manuscript as follows: (3) “. . .and therefore its origin and degree of decomposition. . .”

(1) Line 74: Remove "being". (2) The term will be removed in the revised manuscript.

(1) Line 82: As also pointed out by the other referee a more negative deltaG change would make R2 thermodynamically more favourable. (2) The statement will be corrected in the revised manuscript.

(1) Lines 86-87: Change "are attributed to" to "may arise from". (2) The phrasing will be changed in the revised manuscript.

(1) Line 90: Change "remain" to "remains". (2) The term will be corrected in the revised manuscript.

(1) Lines 92-93 and elsewhere: As also pointed out by the other referee please define each abbreviation when you use it the first time and maintain consistency. (2) All abbreviations will be defined when being used for the first time and only abbreviations will be used afterwards in the revised manuscript. (1) Line 96: Why "to a small extent"? In such shallow systems wind-driven turbulence could disturb the sediments. Lines 97-98: Add "penetration" after "oxygen" and remove "in our case". What do you mean by "perennial circulation". (2) The sentence will be rephrased in the revised manuscript as follows: (3) “. . .but might in the upper parts of the sediments be influenced by oxygen penetration from the water column due to a well-mixed water body.”

(1) Line 99: Please use present indefinite tense, not present continuous. (2) The tense will be corrected in the revised manuscript.

(1) Line 103: "other" sediment properties? (2) The term “other” will be added in the revised manuscript.

(1) Line 108: Change "is accountable" by "accounts". (2) The term will be changed in the revised manuscript.

(1) Lines 110-114: Please rephrase this sentence. (2) The sentence will be rephrased as follows: (3) Until now, laboratory incubations of lake sediments were mostly conducted with samples from one or few sites within one lake with a focus on comparing different lakes with each other rather than covering a high in-lake variability of production rates. Further, these studies emphasize temperature effects on production rates (Duc et al., 2010; Gudasz et al., 2010; Gudasz et al., 2015; Fuchs et al., 2016). Unlike peat soils, where a broad range of controls on CO2 and CH4 production has been investigated, to our knowledge, controls such as organic matter (OM) quality, the occurrence of alternative electron acceptors (EAs), thermodynamic processes and sediment grain size have not, or only individually, been systematically surveyed in small lakes.

(1) Line 119: Did you actually investigate "connected productions patterns to OM"? (2) This sentence might be ambiguous. We will rewrite the sentence as follows: (3) “. . .in

order to relate observed production patterns to measured OM and sediment characteristics, thermodynamics, and water-atmosphere fluxes."

(1) Line 121 and elsewhere: I am not sure if these experiments can be termed as "mesocosm". These were incubations of cores in the lab. (2) To clarify the experimental procedure and the differences between the two laboratory experiments, we changed the descriptions throughout the whole manuscript: Sediment incubations to slurry incubations and Sediment mesocosms to Intact sediment core incubations.

(1) Line 125: Change "hypothesize" to "hypothesized". (2) The term will be corrected in the revised manuscript.

(1) Line 137: Change "blast" to "blasted". (2) The term will be corrected in the revised manuscript.

(1) Line 138: Change "arose" to "formed". (2) The term will be corrected in the revised manuscript.

(1) Figure 1 captions: Technically the depth categories are wrong. For example by <150 cm, you imply depths between 125 and 150 cm, but 20 cm is also <150 cm. This should be clarified (e.g. 125 indicates 100cm<depth_125 cm). (2) The description of lake depths categories will be clarified in the revised manuscript as follows: (3) "...numbers 50, 100, 125 and 150 indicate lake depth category (50: <50 cm, 100: 50-100 cm, 125: 100-125 cm, 150: 125-150 cm)."

(1) Table 1, caption: Analytical procedures do not have to be mentioned here; they should be described in methodology section. (2) The description of analytical procedures will be deleted. These can be found in the methods section 2.4 of the revised manuscript.

(1) Line 167: Change "at three occasions" to "on three occasions". (2) The term will be changed in the revised manuscript.

(1) Line 169: Why randomly? It should be selectively based on a reason. (2) We

randomly chose the sites in order to avoid detecting differences between sites, water depths or transects due to the sampling date. E.g. if we took all four samples from one transect on the same sampling date, we could not have been sure that potentially observed differences in production rates were because of different site characteristics or because of the sampling date. The same would be true for water depths, so we decided to perform a random sampling at each date.

(1) Line 172: Add "respectively" at the end of the sentence. (2) The term will be added in the revised manuscript.

(1) Line 176: Change "added with" to "containing". (2) The term will be changed in the revised manuscript.

(1) Line 180: Change "stored" to "maintained". (2) The term will be changed in the revised manuscript.

(1) Line 192: Referring to a comment by the other reviewer, I note that some isotope data are presented in Table 2 (not for sulphur though), although not at all discussed in the text. It is not clear whether the sample was decalcified. Also what was the reproducibility of measurements? In fact the precision of analysis is not given for any parameter. (2) Isotopic values of C and N will be mentioned in the revised manuscript in the results sections 3.1.1 and 3.1.2. Sulphur isotope data was not measured an will therefore be deleted from the methods section. The samples were not decalcified before analyses. But we analyzed samples for carbonate content which confirmed that carbonate contents were very low (< 0.9 mg/g). We therefore assume that carbonates in the samples only have a minor influence on the results of isotopic data. During every run of samples, multiple working standards were measured to assure reproducibility of measurements. Precision of standards are: < 1% for C, < 0.1 % for N, < 0.05 ‰ for delta13C, < 0.5 ‰ for delta15N. Information on precision will be added to the methods section 2.2.2 of the revised manuscript.

(1) Line 201: Change "Therefore" to "For this purpose". (2) The term will be changed

in the revised manuscript.

(1) Line 253: Something missing in the sentence. (2) The term will be corrected in the revised manuscript.

(1) Line 267: Change "analyzed for" to "measured". (2) The term will be corrected in the revised manuscript.

(1) Lines 274, 291: Change "measured" to "analyzed". (Note samples are analyzed, parameters are measured). (2) The term will be corrected in the revised manuscript.

(1) Line 301: Change "Therefore" to "For this purpose". (2) The term will be corrected in the revised manuscript.

(1) Line 313: These are lab experiments, NOT mesocosms! (2) See comment on line 121.

(1) Line 331: Change "conducted" to "made". (2) Will be changed in the revised manuscript. Please note that the whole sentence should be rephrased in order to the other reviewer's suggestion as follows: (3) "For statistical analyses and discussion, only measurements that were made > 50 days after the deployment of the sediment mesocosms in the climate chamber were used. This was done in order to ensure the system had adapted to experimental conditions and had reached a steady state. Steady state conditions were indicated by quasi-constant CO2 and CH4 concentrations in the sediment."

(1) Line 361: There is no other way to quantify inputs is ebullition? (2) The only way to directly quantify ebullition is via inverse funnels that could trap the emitted methane bubbles. We tested this method in our sediment cores, but without success so that we decided to only measure total methane fluxes and separate diffusive and ebullitive fluxes mathematically as this has been suggested by Bastviken et al. 2004 and adapted by many others when measuring in-situ methane fluxes with a floating chamber approach.

(1) Line 408: Change "nor" to "or". (2) The term will be changed in the revised manuscript.

(1) Fig. 2: Figure difficult to digest. I could not follow "Different letters indicate significant differences between these sites." What do letters "a"-"d" mean? Am I missing anything? (2) For more clarity, the figure will be split into two and production rates in 5-10 cm depth will be displayed in the supporting information. a-d denote if there are significant differences between sites. Same letters mean no significant differences. The description will be rephrased in the revised manuscript as follows: (3) "Identical lowercase letters indicate production rates that were not significantly different (i.e. p > 0.05) from each other."

(1) Line 483: Change "by averagely" to "on a average by". (2) The term will be changed in the revised manuscript.

(1) Lines 490, 493, 499: See earlier comments on Lines 21-22. (2) The sentences will be rephrased according to the above-noted suggestion.

(1) Line 508 and elsewhere: I believe sediment ebbulition in inferred from k>o. I am not sure. Was there any bioturbation that could increase the emission? (2) Ebullition was inferred from piston velocity k > 2 as described in the methods section 2.3 (see lines 359-361 of the submitted manuscript). We did not observe any bioturbation during the experiment.

(1) Table 4: "n.s." presumably means not significant (p<0.05). Is is mentioned somewhere? Significance also depends on the number of values that are not given. (2) n.s. means not significant. An explanation will be added to the table's description as follows: (3) "n.s. means that correlations were not significant (p > 0.05)." We will revise the table including number of values as follows: CH4 flux CO2 flux rho p n rho p n Clay 0.648 < 0.05 12 0.605 < 0.05 12 Silt 0.497 n.s. 12 0.302 n.s. 12 Sand -0.648 < 0.05 12 -0.605 < 0.05 12 Fats, waxes, lipids -0.833 < 0.05 8 -0.333 n.s. 8 Phenols; humics -0.833 < 0.05 8 -0.357 n.s. 8 Aromates -0.595 n.s. 8 -0.524 n.s. 8 Lignin -0.786 < 0.05

8 -0.381 n.s. 8 C/N -0.881 < 0.01 8 -0.333 n.s. 8 C (%) -0.714 n.s. 8 -0.190 n.s. 8 CH4 sediment stock change -0.222 n.s. 41 0.05 n.s. 35 CO2 sediment stock change -0.049 n.s. 41 -0.064 n.s. 35

(2) Numbers of values for other calculated correlations will be given in the Supporting information Tab. S1.

(1) Lines 528: Change "concentration" to "concentrations" and "was" to "were". (2) The term will be changed in the revised manuscript.

(1) Line 536: Change "were significantly negative correlated" to "showed significant negative correlation" (2) The term will be changed in the revised manuscript.

(1) Line 542: What do you mean by "narrower"? lower? (2) Narrower will be changed to lower in the revised manuscript.

(1) Lines 545-556: Authors have emphasized on C/N ratio. They have observed increase in C/N with depth in the inner part of the lake. C/N ratio may not be a very efficient parameter to characterize organic source owing to rapid remobilization of nitrogen as well as reabsorption of ammonium on particulates. The paragraph 405 “The C content in the samples was between 2.15 and 33.16% with lowest values at site 3.50 and highest at site 1.50. C/N ratios ranged from 10.97 at site 1.150 to 19.06 at site 3.100. Neither C content nor C/N ratio showed significant changes with sediment nor lake depth, but C/N ratio was significantly higher in samples taken close to the shore (50) than in samples from the lake center (150) ($p < 0.01$).” is very confusing. A graph showing distribution of C/N ratio across the horizontal length of the lake would suitable to comprehend the results better. (2) We will include a figure showing C/N ratios and absorption ratios for fats/polysaccharides to better illustrate our results. (3)

(1) Line 551-552: I do not believe in shallow depths it matters. (2) We additionally propose the mechanism of resuspension and focusing of small particles, that could alter the degree of decomposition of OM. (3) “. . .As this process might not be of the

same importance in shallow lakes compared to deeper lakes, we additionally suggest that the more decomposed OM in the lake center might have undergone degradation processes during resuspension and focusing of small particles as a result of wind-induced bed-shearing (Mackay et al., 2017).”

(1) Line 559: Change "buried" to "getting buried" (2) The term will be changed in the revised manuscript.

(1) Line 571: Change "role for" to "role in" (2) The term will be changed in the revised manuscript.

(1) Line 578: Change "e.g." to "among other things" (2) The term will be changed in the revised manuscript.

(1) Line 587: Change "in the following" to "below" (2) The term will be changed in the revised manuscript.

(1) Line 591: Remove "the" before "in other studies" (2) The term will be removed in the revised manuscript.

(1) Lines 612-614: Laborious sentence. All you are saying is that such shallow depths do not get thermally stratified in summer. (2) We will rewrite the sentence as follows: (3) “. . .especially regarding the fact that shallow waters, as against deeper lakes, do not get thermally stratified in summer and therefore shallow sediment warm much faster (Jankowski et al., 2006).”

(1) Lines 619-620: Change the tense to present indefinite. (2) The term will be changed in the revised manuscript.

(1) Line 627-630: What do you mean by "wider" and "narrower"? I do not follow this sentence. (2) “Wider” and “narrower” will be changed to “higher” and “lower”. We will restructure the whole discussions part about OM quality and CO2 and CH4 production rates according to the other reviewer’s suggestions. To explain what we mean by this statement: C/N ratios can be interpreted in two ways: a) high C/N ratios = low decom-

position state, low C/N ratios = high decomposition state; b) but C/N ratio can also be used to differentiate between OM of terrestrial and aquatic origin (see Meyers 1994) whereas high ratios = terrestrial and low ratios = aquatic origin. It is known that OM of a low decomposition state is easier degradable for microorganisms and therefore leads to higher production rates of CO2 and CH4, but on the other hand, aquatic OM is usually easier degradable for aquatic microorganisms and would therefore lead to higher production rates compared to OM of terrestrial origin (see e.g. Grasset et al. 2018). We therefore conclude that, although there exist two contradicting effects (low vs. high decomposition or aquatic vs. terrestrial origin), the fact that OM closer to the shore is in a lower decomposition state (although it is probably of terrestrial origin) fuels CO2 and CH4 production. The paragraph on C/N ratio and production rates will be revised as follows: (3) "C/N ratios are frequently used to characterize the degradation state of OM, but we did not find correlations between C/N ratios and CO2 and CH4 production rates in the slurry incubations. Although OM of autochthonous origin was found to fuel higher degradation rates than allochthonous OM (West et al., 2012; Grasset et al., 2018) we found evidence of predominant inputs of allochthonous (terrestrial) material at sites with higher production rates close to the shore (higher C/N ratios), whereas sites with lower production rates in the lake center received mainly autochthonous (aquatic) OM as indicated by lower C/N ratios (Meyers, 1994). On the other hand, high C/N ratios also indicate a lower degradation state and therefore higher degradation potential whereas low C/N ratios are usually typical of highly decomposed OM having a lower CO2 and CH4 production potential (Malmer and Holm, 1984; Kuhry and Vitt, 1996). These two possibilities of interpreting C/N ratios might be the reason for apparently contradicting findings and the missing relationship between C/N ratios and CO2 and CH4 production rates."

(1) Line 637: Change the tense to present indefinite. (2) The term will be changed in the revised manuscript.

(1) Line 654: But the acetate concentration increased! (2) Please not the discussion in

section 4.1.4 of this observation in lines 660-665 of the submitted manuscript.

(1) Line 655: Remove "of" before "importance" (2) The term will be changed in the revised manuscript.

(1) Line 671: OM quality is not quantified so instead of low you should perhaps use poor. (2) The term will be changed in the revised manuscript.

(1) Line 673: Change "of energy" to "in energy" (2) The term will be changed in the revised manuscript.

(1) Line 675: Change "... acetate, but rather is fermentation" to " ... acetate. Instead fermentation may be rate limiting" (2) The term will be changed in the revised manuscript.

(1) Line 677: Bring "Further" before "it". (2) The term will be changed in the revised manuscript.

(1) Line 678: Change "finding emphasizes" to "supports" (2) The term will be changed in the revised manuscript.

(1) Line 692: If the relationship was insignificant the trend cannot be "clear". (2) The term will be removed in the revised manuscript.

(1) Line 693: Not at all clear, and so is the following conclusion. I find this whole paragraph speculative. (2) We will rewrite the whole paragraph 4.1.5 - also following the other reviewer's suggestions – where we will elaborate the relationships between CO2 and CH4 production and alternative EAs more precisely. Instead of discussing relationships between EAC and CH4 production, we emphasize that measured inorganic and organic EAs can explain 40-80% of measured CO2 production. The missing capacity can probably be explained by solid-phase iron, which we found ranging from 2 to 3 %, but whereof we do not have information on its speciation. We further emphasize, that missing correlations between EAC and CH4 production are due to our experimental set-up: the one-week pre-incubation might have already depleted a large amount of

reducible organic and inorganic EAs so that subsequent changes and therefore correlations were low.

(1) Line 702: Change "something" to "somewhat" (2) The term will be changed in the revised manuscript.

(1) Line 706: Change "approaches" to "factors" (2) The term will be changed in the revised manuscript.

(1) Line 730: Authors attempt to correlate ebullition with grain size. They believe that higher sand content leads to lesser ebullition. Which is highly unlikely since ebullition depends on permeability of sediments and not porosity. Sand always has higher permeability than silt and clay although lesser porosity. You need to elaborate your concept with more clarity (2) When explaining CH4 ebullition with the concept of grain size distribution/porosity, it is not primarily of importance how permeable the material is, but how effective bubbles can actually accumulate in the sediment (so that they can subsequently be released). Lui et al. 2018 found that the dominant pathway of bubble formation is by displacing the surrounding sediment, and that this is easier in soft, silty sediment compared to sandy sediments. This sediment displacement would lead to more macropores and therefore a higher connectivity creating conduits for bubble release. We will change the paragraph explaining these mechanisms more precisely as follows: (3) “We found ebullition supporting significantly to total CH4 fluxes in two of our four intact sediment core incubations, whereas sites with higher shares of sand exhibited less ebullitive fluxes confirming the findings of Liu et al. (2016) and (2018). The authors explain their findings with the dominant pathway of bubble formation in the sediment, which is by displacing surrounding sediment particles. As this mechanism is more efficient in soft silty sediments compared to sandy material, CH4 bubbles likely accumulate more easily in silt, creating a network of macropores and therefore conduits for subsequent bubble release. We further found OM quality partly exhibiting significant negative correlations with CH4 fluxes, but to a lesser extent than with CH4 production. When preparing slurry incubations, the physical sediment structure is de-

stroyed, so that OM quality becomes the major controlling factor for gas production. These findings suggest that grain size distribution is besides OM quality a main driver of spatial CH4 flux patterns in intact sediment core incubations and that only a combination of physical characteristics and sediment OM quality could sufficiently explain CH4 emission patterns from lakes.”

(1) Line 747: Change "experiment" to "results" (2) The term will be changed in the revised manuscript.

(1) Line 749: Remove "especially" (2) The term will be changed in the revised manuscript.

(1) Line 753: Change "vulnerable" to "sensitive" (2) The term will be changed in the revised manuscript.

(1) Line 754: Change "unroll" to "expect" (2) The term will be changed in the revised manuscript.

(1) Line 755: Change "lower water columns" to "shallow depths" (2) The term will be changed in the revised manuscript.

(1) Line 761: Change "refer" to "attribute" (2) The term will be changed in the revised manuscript.

(1) Lines 764-765: Then why do you not find strong relationship between methane production and (EACorg)? (2) Please see also comment on Line 693. The statement will be discussed in more detail in the revised manuscript.

(1) Line 770: Measuring "production rate" does not neglect water column processes, interpretation of these data alone would. (2) The sentence will be rephrased in the revised manuscript as follows: (3) “Further, measuring production rates only would neglect the importance of the water column as a sink of sediment generated CH4. . .”

Please also note the supplement to this comment:

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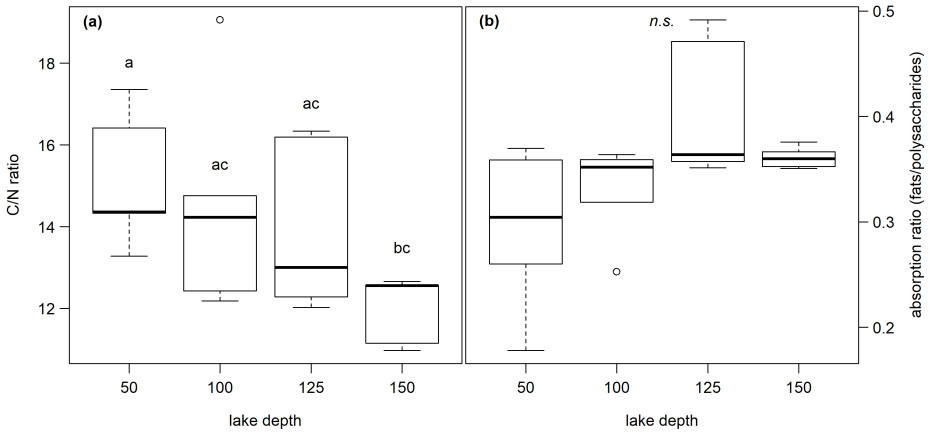

Fig. 1. C/N and FTIR peak ratio vs. lake depth category (see comment on ll. 545-556)