

***Interactive comment on “Microbial
methanogenesis in the sulfate-reducing zone in
sediments from Eckernförde Bay, SW Baltic Sea”
by Johanna Maltby et al.***

Johanna Maltby et al.

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Received and published: 14 September 2017

We would like to thank the reviewer for her/his critical comments, which we think helped to improve the quality and clarity of this manuscript. We hope our responses and adaptations are adequate to accept this manuscript for publication in Biogeosciences. Please find our detailed responses below. Anonymous Referee #2

Received and published: 15 June 2017

The work presented by Maltby et al. is really nice piece of study gathering results from several impressive campaign of sampling and involving different cutting-edge methods.

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Their findings give an interesting overview of biological processes and environmental factors controlling methane emissions from sediments and water column of a Baltic sea bay, well known for its importance in global methane emissions. The originality of their work lies in the demonstration of co-occurrence of sulphate reduction and methanogenesis in surface sediments. This co-existence is permitted by a mechanism developed by some methanogenesis microorganisms to escape from the strong competition with sulphate-reducing microorganisms: using (releasing? I did not find information on that) non-competitive substrates. The manuscript is overall well written except the abstract, see my comments below. I have only two main concerns.

First, the article is sometime written in a way that only initiates of the field may touch. The first sentence of abstract directly starts with the work done without putting the study in a wider context. The object you study is complex and well structured. We do not immediately understand the relevance of studying methanogenesis in the sulphatereducing zone.

Authors Reply: We thank the reviewer for this comment. We agree that the abstract starts abrupt and therefore included a short introduction to explain the reasoning for this study.

We neither understand that you studied surface, deep sediment and the water column and not only surface (sulphate-reducing) sediment. The reference of “a non-competitive substrate” is not understandable. Which competition do you refer ? Implying which organisms?

Authors Reply: We hope that these questions are answered in the abstract now after we added some introduction and definitions.

In the introduction, it could be useful to build a synthetic figure summarizing the studied ecosystem including the different compartments, different organisms, interactions among organisms, exchanges of matters between these compartments.

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Authors Reply: We appreciate this idea and added a scheme (see new Fig. 1).

Second, the (minor) contribution of surface methanogenesis to total methane emissions from this ecosystem is a bit hidden in the article. This contribution deserves to be clearly presented in the abstract. To my point of view, the minor contribution of this mechanism does not question the quality and relevance of this study, and is an important information.

Authors Reply: We understand that the role of surface methanogenesis to total benthic methane emissions is not discussed in length in this paper. However, to be able to go into more detail about the contribution of surface methanogenesis, more detailed studies about emission rates into the water column, methane oxidation rates in the sediment (oxic and anoxic) as well as in the water column would be necessary. Our presented data set gives a first idea about surface methanogenesis and its potential role, but anything beyond that would be speculative.

In the same vein, the statement that surface methanogenesis could play a key role in fueling the surface anaerobic oxidation of methane is speculative since this last process was not measured in the study.

Authors Reply: We agree that the statement is speculative, as AOM was not determined in the present study. But we have good indications of surface AOM from radio-tracer incubations from previous studies (Treude et al. 2005) in the Eckernfoerde Bay. Furthermore, we see no issue with formulating a hypothesis.

Specific comments. Line 30 suppress "in the manipulated experiments.

Authors Reply: Done.

L31-33 this new objective that pops up too late. Please gather your objective in one sentence

Authors Reply: We gathered the objectives.

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L47 replace “makes an important contribute” by “substantially contributes to”. I did not understand the last part “as it could...”

Authors Reply: We changed the sentence to make it clearer.

L78-79 and throughout the manuscript. The expression “Environmental control mechanisms” is a bit elusive. Do you mean “environmental controls” or “biological processes”? Try to better specify what should be better studied.

Authors Reply: We changed it to environmental controls wherever we used “environmental control mechanisms”.

L164 Rewrite your sentence to clarify. Could be “Biological activities of samples were stopped by the addition of mercury chloride solution...”

Authors Reply: Done.

L177 “extracted”? you mean “sampled” for analysis?

Authors Reply: "porewater extraction" is a commonly used expression in sediment geochemistry.

L193-216 I am not expert in measurement of methane concentration in sediment, but I am wondering whether the fact of cutting sediment core in 1 cm sediment interval could release, at least a part of, the methane you wish to quantify.

Authors Reply: It is correct that some methane in form of gas bubbles might escape during the slicing procedure as it does during multicoring. However, we accept that fact of potential methane loss, as these gas bubbles originate from deep methanogenesis below the sulfate-reducing zone. We are more interested in the methane kept in the tiny spaces between the shallow sediment, which we indeed can determine with the presented method.

L236 Could you rapidly explain again what is the hydrogenotrophic methanogenesis? And what is the interest of measuring this in the context of your study?

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Authors Reply: Done.

L389 Suppress “in” before september.

Authors Reply: Done.

L401 I guess you’re talking about the C/N of particulate organic matter, but I am not sure. Please specify.

Authors Reply: Done.

L543 I propose you to replace the end of your sentence by “...the following observations that will be discussed in more detail in the following chapters”. My first reaction was to try to understand your arguments before reading the following chapters.

Authors Reply: Done. Thanks for pointing that out.

L569 Your explanations about the competition between sulphate-reducing and methanogenesis microorganisms, and the strategy of methylotrophic methanogens to escape from this competition, are very clear and convincing. Now I am wondering whether there is competition between hydrogenotrophic and methylotrophic microorganisms. And if yes, does this competition change with depth?

Authors Reply: To our knowledge, there is no competition between hydrogenotrophic and methylotrophic methanogenesis, as these metabolism are using different substrates (that is why this categorization makes sense). Some hydrogenotrophic methanogens are able to use secondary alcohols in addition, however, they normally cannot use the substrates of methylotrophic methanogens (methanol, methylated amines, dimethylsulfides).

L587-592 This sentence is too long. Split your explanations into 2 sentences.

Authors Reply: Done.

L614 If I follow well, you should add a “P” after “DMS”.

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Authors Reply: Done.

L605-631 Maybe this is a limit of your study of not having quantified some key non-competitive substrates in sediments and water. It could be discussed in a paragraph drawing next investigations that could be done.

Authors Reply: We agree with the reviewer that this is a limit of our study. Our focus in the present study was to see the potential rates when fueled with non-competitive substrates such as methanol. The next step would be to measure the natural concentrations. We added a paragraph at the end of 4.1.4 to advise the reader that we actually did not measure most of the non-competitive substrates (besides methanol in June 2014).

L640 What fractionation are you discussing? An isotopic fractionation? You must better explain.

Authors Reply: Changed to isotopic fractionation.

L488-489 Could you check whether such moderate isotopic fractionation (factor of 1.07-1.08) could explain an increase of delta of almost 200 per mille. I have a doubt.

Authors Reply: We used fractionation factors from previous studies using non-¹³C-spiked methanol. With our data set we were not able to calculate a reliable fractionation factor, as you have to use concentrations of substrate and product from the stationary phase. However, the methanol in our experiments was used up before a stationary phase was observed (see Fig. 6 (now 7). At timepoint 2 (14 days) the fractionation factor was 1.22 (calculated by authors), but this value should not be used, as it was retrieved from the slope phase. To actually be able to determine the fractionation factor in our case, more experiments would have to be conducted.

L644-645 This sentence is not clear. What would be the alternative explanation?

Authors Reply: Revised the sentence, as there is no other explanation.

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L646 One bracket is lacking at the end of sentence.

Authors Reply: Done.

L684-694 I did not understand your explanations. Please try to reformulate and be more direct when you propose an interpretation.

Authors Reply: Revised the sentences.

L706-707 Did you find results going in this way as well?

Authors Reply: As described in this paragraph, the PCA did indeed show a positive correlation between methanogenesis and organic carbon availability, supporting the idea that organic matter availability can lead to coexistence of sulfate reduction and methanogenesis. To really answer your question, we would also need results from less-organic-rich systems, such as open ocean sediments.

L713-736 This section is really too long. Split it in two paragraphs, one focusing on the effect of POC amount and the other on C/N ratio.

Authors Reply: As POC and C/N are directly interlinked, we would prefer to discuss both factors together. We think it is necessary to discuss the availability of POC and the freshness of POC together. However, we formulated subtitles to make this paragraph easier to read.

L830-831 I do not understand your interpretation of the positive correlation between surface methanogenesis and C/N ratio of POM.

Authors Reply: We changed the sentence a little bit to make it more clear. Essentially, a positive correlation means higher methanogenesis when C/N ratio is higher, thus indicating less-fresh material. Usually, with increasing sediment depth the organic material becomes less-fresh (= higher C/N ratio). But at these depths, fermentation processes take place, which then provide substrates to the methanogens. So it is not surprising that methanogenesis is high at deeper depth.

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L805-809 and 832-834. This process of anaerobic consumption of methane (AOM) was not measured in this study making all these discussions around the key role of surface methanogenesis in fuelling AOM very speculative. I do not understand why deep methanogenesis, which contributes for the major part of methane emissions, does not contribute to AOM fueling. It sounds like you would absolutely like to give a central importance to surface methanogenesis.

Authors Reply: We agree that this is a speculative statement. But we are confident making this connection between surface methanogenesis and surface AOM, as it was measured in similar sediments from the same sampling area. However, we softened our conclusion and provided more details for our reasoning.

Please also note the supplement to this comment:

<https://www.biogeosciences-discuss.net/bg-2017-36/bg-2017-36-AC2-supplement.pdf>

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2017-36>, 2017.

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