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Interactive comment on “Timescales for the development of methanogenesis and free gas layers in recently-deposited sediments of Arkona Basin (Baltic Sea)” by J. M. Mogollón et al.

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1 Introduction

We greatly appreciate the constructive comments posted by Prof. Dickens and the anonymous reviewer, especially since these suggestions will likely make the manuscript more appealing to a broader audience. We have also attempted to address all of their concerns. Below are the original major and minor comments posted by the reviewers (*italics*) as well as a detailed response to each point (**bold**). Note also that the reference included only pertain to the response and not to the reviewers’

Full Screen / Esc

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Discussion Paper



comments.

2 Reply to Prof. Gerald Dickens

Mogollón and colleagues present model simulations to address an intriguing question: how does methane accumulation proceed over time on the continental shelf? To my knowledge, the problem has been discussed rarely in the literature from a dynamic perspective, let alone modeled. I found the work interesting and insightful. I enjoyed reading the manuscript, and it makes one think. The model also contains some important concepts missing in quite a bit of the literature (e.g., the rising of SMT depth and the relative importance of AOM with increasing methane production over time). I have no major criticisms of the work. However, I do think they could enhance the work, so that it becomes more interesting to a broader community. I also have a series of minor comments. Lastly, I offer an apology for the lateness of my review, even though I was not asked to review the manuscript until October 31. I trust the comments are fair and constructive.

Main Comments

(1) The "big picture" and, ultimately, the reason for the manuscript could be articulated much better. Right now, the main rationale for the work seems to be that rates of methane generation are an important and open issue in our understanding of methane and carbon cycles (p. 7625; Lines 13-15). This is not obvious, especially because the concept is not succinctly stated in the given reference (Reeberg, 2007), and because very few carbon cycle models include methane in marine sediment and relevant fluxes. Moreover, the paper discusses methane amounts, as well as methane outputs. The manuscript (and certain sections) would become stronger if they rewrote the Introduction so as to place the totality of their work in perspective (see also Comments below). With some liberty, here's the overall problem I think they are chasing:

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

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Discussion Paper



- Enormous amounts of methane occur in marine sediment, on both continental shelves and continental slopes - This methane is dynamic with defined carbon fluxes to and from the ocean - A broad array of evidence suggests that the masses and fluxes of methane in seafloor sediment can vary significantly over time - For example, on the shelf, there are features indicative of gas expulsion (e.g., pockmarks; e.g., numerous papers by Hovland and Judd) or remnants of methanogenesis where none exists today (e.g., ^{13}C -enriched authigenic carbonate; e.g., Malone et al., Mar. Geol., 2002) - However, the evolution of methane systems remains poorly constrained

We agree with Prof. Dickens regarding the lack of a big picture in the introduction. We have thus taken his suggestion into account and rewritten the first paragraph in the introduction.

(2) A series of models have been developed for the evolution of methane systems on continental slopes (e.g., Davie and Buffett, JGR, 2001, Chatterjee et al., JGR, 2011 and several others in the intervening time). While these modeling efforts have some similarities to the current one (and its predecessors), they also have some notable differences. There is no need to delve into these models in detail, given the focus of the current effort on the shelf. However, right now, there is no mention of these models or of methane cycling on the slope in general. The potential problem here is that the current effort may perpetuate several misconceptions about seafloor methane in general; more specifically, some of the results do not apply to seafloor methane cycling in deeper water, and this should be stated.

For example: - Sites on the slope also have horizons defined by methane solubility; however, gas hydrate can form, which changes the phase relationships considerably. - Sites on the slope are not subject to sub-aerial exposure and "flushing" over glacial-interglacial cycles; consequently, and with the introduction of a solid methane component, enormous amounts of methane can build-up over millions of years. - Sites on the slope almost invariably have low temperatures on the seafloor; as such, the temperature rise with depth becomes important to methanogenesis. - Sites on the slope

generally receive much greater organic carbon during glacials; this is opposite to the shelf. A very intriguing concept derives from the present work and efforts on the slope: the average depth of total methane production might vary considerably over time; that is, during interglacials, considerable methane production might occur on the shelf at the expense of methane production on the slope.

We agree with the reviewer's point. This is the main rationale why we restricted our title, introduction, and discussion only to Arkona Basin and possibly other shelf sediments. In an attempt to broaden the scope of the paper as well as to distinct clearly methane dynamics in slope sediments from those of the shelf, we have modified the introduction and conclusions.

(3) As stated above, the modeling provides some interesting perspectives on methane cycling in sediment on continental shelves. However, this is not discussed in a general sense. Above and beyond any comparisons to slope environments, their modeling may apply to other shelf environments. Although many shelves do not contain a lacustrine deposit, Holocene sections are often underlain by a hiatus emplaced during sea level lowstand and sub-aerial exposure, which may mark a time-zero for methanogenesis. I suspect their modeling provides a broader framework than presented.

We agree that this approach may be suitable in similar shelf environments and have clarified these aspects in the introduction and conclusion.

(4) There are some clear tests of their modeling. For example, there should be very specific profiles for DIC (alkalinity) and the ^{13}C of this DIC (e.g., Chatterjee et al., JGR, 2011). It would be good to state this.

In this study we use methane, sulfate, POC concentrations and sulfate reduction rates to constrain the rate of POC mineralization and AOM. The rates model could be further verified using DIC, alkalinity, ^{13}C -methane, ^{13}C -DIC distributions (e.g. Martens et al., 1999).

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Obviously the modeling would benefit from isotope data, yet they were not measured at our site.

(5) Temperature is discussed as a key parameter. There is also commentary on how temperature changes with seasons. However, other than a "reference temperature", values are never stated. It would be good to explicitly state temperatures and their ranges in the text (place in parentheses). To a lesser degree, the same applies for pressure and salinity.

Temperature values are stated in figure 3. We have expanded the methods section to include further information on temperature including the governing function and the parameters used to calculate temperature (Table 2).

Specific Commentary

- Page 7625 -

Line 5: The "tense" is mixed with the phrase "becomes deposited".

The tense has been fixed to "became deposited".

Lines 15-18: The writing seems to suggest that methane is particularly abundant in sediment under hypoxic water masses. This generally may be correct for sediment on the shelf; obviously, however, enormous amounts of methane can occur in sediment on continental slopes, which generally do not underlie hypoxic water masses. (See also Comments 1 and 2).

This sentence was rewritten to pertain only to shelf sediments. We have also clearly distinguished the slope dynamics from the shelf (comment 1). There should thus be no possible confusion anymore.

- Page 7626 -

Line 1: Of course, methane can also be consumed aerobically, when it escapes to very shallow sediment and the water column.

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



The sentence was changed to: "... can be consumed either aerobically in the presence of oxygen, and/or more commonly, by anaerobic oxidation of methane..."

Line 5: I am not sure why SMTZ is being used instead of SMT as in many papers (transition and zone seem redundant to me).

The authors were following the convention of previous studies and papers. We prefer to adhere to this convention.

Line 6: This is correct with the caveat of venting (which can include dissolved gas and free gas bubbles).

We only refer to methane escape in the aqueous phase on passive margins. We have modified the text accordingly.

Lines 7-10: This sentence (and concept) is not clearly presented. This is because the idea of saturation zones that relate to methane concentration and methane flux is never presented. This might be addressed with a new Figure 1 that shows the generalities of what they are trying to model (i.e., a figure showing generic methane and sulfate concentrations versus depth).

This sentence was not clear. We have now removed it, rephrased the previous sentence and added references where these concepts are clearly explained.

Lines 15-18: I do not follow the glacial-interglacial reference, as the model and paper strictly pertains to the Holocene. Moreover, others have certainly discussed the potential long-term impact of AOM on geochemical cycles (e.g., Dickens, EPSL, 2003; Dickens, Clim. Past, 2011).

We have disregarded the glacial-interglacial reference and replaced it with "... over millennial timescales...". We were referring to modeling studies, and have now explicitly stated it: "Furthermore, few modeling studies..."

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

- Page 7627 -

Lines 7-8: Is this really why stratification occurs? I thought it also reflects the large freshwater input to the Baltic Sea.

The sentence has been rewritten to account for the reviewer's valid point: "... stratification of the water column due to both dense saline waters entering from the North Sea and increase in river runoff during glacial melting, coupled to a rise in primary productivity..."

Line 14: Missing a word(s).

We have added "of" which was missing between "absence" and "deep"

Line 19: As noted below, it is not clear whether the low organic carbon in the lower sedimentary units refers to TOC or reactive organic carbon.

The low organic matter refers to the lake sediment. The sentence was modified to account for this change: "... HORMs. A lacustrine sediment with no organic carbon or sulfate and methane in the porewater represents the initial condition for the model."

- Page 7628 -

Line 2 (and throughout): Remove "the" for proper nouns (nb. this is usually the case in the present manuscript but not always).

"the" was removed when preceding Arkona Basin. However, "the" was maintained when referencing the Baltic Sea and the North Sea.

Line 5: Rewrite. Maybe I am wrong, but I think that it's the seafloor of the basin that is 50 m deep.

The reviewer is correct and we have thus added "water" to the sentence.

Line 8: The use of "while" is uncertain here because the gas horizon and fluffy layer

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8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



do not seem conflicting geographically.

"While" has been replaced with "and".

Lines 10-11 (and throughout): I would rewrite so as to make the sentence active instead of passive. "Significant : : : punctuated : : :" (This obviously stylistic but, in my opinion, makes reading much smoother and better).

This sentence was changed to an active voice.

Line 10 and onwards: This is a very long paragraph with multiple concepts. I would split to make reading easier.

The paragraph was separated into three different paragraphs which illustrate the different concepts.

- Page 7629 -

Line 8: This is not clear because usually sediments are described downcore (i.e., with increasing depth). Here, however, it seems to be upcore and with decreasing depth.

The reviewer is correct. We have thus changed the description to "... since it is characterized by an increase in the concentration of particulate organic carbon (POC) as well as a change from light-dark laminated gray sediments to a dark grayish-green sediment with decreasing sediment depth ..."

Lines 11-12: Fix the formatting here in regards to the referencing.

The sentence was rewritten as follows: "In this study, we assume that the initial HORM deposition in the southwestern Baltic Sea began 8.0 kyr BP ..."

- Page 7630 -

Line 1: Do the "missing tops" pertain to all cores or gravity cores? I assume the latter, but this is not clear.

Inserted the word "gravity" into the sentence.

C6155

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



Lines 6-8: I would add two sentences here briefly stating how the measurements were made. There is no need for detail if documented elsewhere; however, one should not have to read other works to know the basics.

More information about the measurements has been added.

Line 18: It should be stated that the second reaction (methanogenesis) is an abbreviated reaction. In other words, this reaction does not exist per se; rather it is a summary of intermediate reactions.

The word "net" has been added to the sentence to account for the fact that these represent overall reactions which sum the intermediate pathways for organic matter hydrolysis and fermentation.

- Page 7632 -

Line 17: Is alpha-o always zero? If not, why?

α_0 may not necessarily represent a value of zero. For the purposes of inert sediment grains and the bulk sediment, α_0 is approximately zero in Arkona Basin.

- Page 7633 -

Line 4: The word "impressed" should be changed.

The word impressed is used in the classical works of sediment diagenesis (e.g. Berner, 1980; Boudreau, 1997, and references therein). We therefore adhere to this terminology.

Line 11: First, clarify this with "downward" in regards to advection of free gas. More importantly, I would add a caveat. This assumption effectively means that free gas is kept below some threshold concentration. Presumably, over time, sufficient free gas could accumulate, such that this assumption is not longer valid and the model would have to be amended.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



We discuss this caveat in the discussions section. Studies on bubble migration have demonstrated that single bubbles may fracture the sediments provided the bubble pressure exceeds the effective stress of the sediment. Likewise, the bubble may migrate through the pore space, provided that buoyancy forces can overcome the capillary pressures. No threshold value has been determined in relation to bulk gas content (at least, to the knowledge of the authors). Since adapting an unknown threshold value would add too much speculation to the present model, we instead have added the caveat that the model provides an upper bound for the methane gas inventory.

Lines 15-25: I think there is an intrinsic assumption here: namely that degradation of organic carbon through R1 and R2 proceeds similarly. In other words, the same suite of compounds is used for sulfate reduction of POC and methanogenesis. They may want to comment on this issue.

The reviewer is correct. We assume that sulfate reduction and methanogenesis are not rate limiting in the degradation of organic matter, but rather that extracellular hydrolysis is the rate limiting step, a common assumption for these models (e.g. Westrich, 1983). We have added the following caveat to the manuscript. "Assuming extracellular hydrolysis to be the rate-limiting step controlling the redox geochemistry, we ..."

- Pages 7634/7635 - Given somewhat similar modeling of methane accumulation on continental slopes, it may be worth noting that, at the water depths and pressure in the Baltic Sea, gas hydrate cannot form, so this phase, which complicates matters significantly, can be ignored in the modeling.

We have added the following sentence to address this issue: "Note that, given the pressure, temperature and salinity regime in Arkona Basin sediments, methane hydrate cannot form and is therefore excluded from the model."

- Pages 7636 -

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Lines 4-6: Does the 5% TOC refer to sediment below the HORM? Currently, these lines read this way, but this contradicts statements elsewhere. If so, this assumption leads to an interesting question: why is this organic carbon unreactive? Has it been subaerially exposed? (I cannot tell from the "history", above). It would be helpful to have typical TOC ranges presented in Figure 2.

We have rewritten the sentence to provide a better justification for not including POC and methane in the lacustrine sediment: "The present stations did not penetrate into the Ancyclus Lake stage. Nevertheless, other cores from Arkona Basin have shown that Ancyclus Lake sediments have a light gray color typical of subaerial exposure Kortekaas et al. (2007)), have a significantly smaller organic matter content with respect to the Littorina Sea stage (Sohlenius et al., 1996), and, in several intervals, are even characterized by organic-free sands (Jensen et al., 1999). Thus ..."

-7637 -

Line 6: awkward referencing

The sentence was changed to reflect that our chloride and sulfate bottom-water variations are based on the salinity curve estimated by Gustafsson and Westman (2002).

-7639 -

not clear

Paragraph was modified to make more readable.

- Page 7641 - Line 12: Change to "across the basin"

Change has been made.

Line 15: This is awkward. Change to ": : : AOM, driven by upwards diffusion of methane generated : : :"

This change has been implemented.

Line 21: Change "evaluate" to "predict".

This change has been implemented.

The writing on this page is a bit unclear with regards to methane saturation. I assume that the 2mM measured on ship is the solubility of methane at 0.1 MPa and shipboard temperature of XX C, whereas those modeled are at XX MPa and sediment temperature of XX C. A bit of information and explanation would be helpful, especially to the casual reader.

The text already mentioned that degassing takes place once the core is retrieved to ambient pressures. This has been clarified.

- Page 7642 -

Line 2: It would be helpful to express this in terms of pore space volume, rather than bulk sediment.

We have added this information.

Line 8: This is confusing because the manuscript seems to state that these sediments have high organic carbon content (p. 7636, Lines 4-6).

We have revised the manuscript to clarify that the limnic sediments do not contain significant amounts of organic carbon.

- Page 7643/7644 -

This section is difficult to follow given the current figures. Figure 7 does not explicitly show a deep SMT; the curves on Figure 9 are not easy to tell apart.

A new panel in Figure 7 has been added to show the double SMTZ. And the panels in Figures 5, 6, 7, and 9 have been switched to color to help distinguish the curves.

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



- Page 7644 -

Lines 5-6: Another very good example of a double SMT occurs on the Peru shelf (D'Hondt et al., Science, 2004). Here, however, it is because a sulfate-rich brine lies in deeper sediment.

We agree with the reviewer that double SMTs occur in other settings. However, we exclude them from the discussion since their depositional/tectonic environment and local geochemistry are very different to those in our study.

Line 13: Is this not for the upper SMT?

The word shallow was added to distinguish the top SMTZ.

Lines 22-24: Is the discussion of temperature correct? It seems to me that this would depend on the change in bottom water temperature, time and sediment properties. Basically, how do heat variations propagate into shallow sediment (See also Comment 5).

The paragraph on temperature has been expanded to include the governing equation.

- Page 7645 -

Line 5: The notion of methane escape is not clear as presently written. Does this refer to ebullition and venting, which is not part of the model? Or does this refer to greater AOM through a steeper pore water profile?

It refers to both. The main point is that a lower solubility can lead to more free gas production, which may increase the likelihood of gas escape from the sediment surface.

Line 22: I do not follow "and the sulfate diffusion." Does this mean the downward flux of sulfate through diffusion?

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



Correct, we have thus changed the sentence accordingly.

- Page 7646 -

Line 11: Does the 10-15% hold despite potential free gas accumulation? It seems to me that this might increase once free gas begins to accumulate.

Correct, there is an increase with free gas accumulation. The increase, however, is within the 10-15% range.

- Page 7647 -

Line 11: Change "considerably flat as compared" to "relatively invariant compared".

Change has been implemented.

- Page 7648 -

Line 17: Which high fractions? Methanogenesis? Sulfate reduction of POC? Both?

Methanogenesis, which was added to the sentence in order to avoid confusion.

- Page 7660 -

Figure 3: I am uncertain what shoreline displacement means. More crucially, the vector is not clear (does this mean positive or negative sea-level?). A second curve on this panel - seafloor pressure - would be helpful.

Shoreline displacement is the term given by the authors (Bennike and Jensen, 1998) to the paleo sea-level fluctuation. We have added to the caption that the curve in question represents the sea-level drop with respect to contemporary levels.

- Page 7661 -

Figure 4: It would be helpful to have a horizontal line (or better yet shaded boxes) marking the HORM and "pre-HORM".

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



The entire figure falls within the HORM sequence.

- Pages 7662/7663 -

Figure 5: It may be the figure copy and the distinction between solid and dotted lines, but something seems incorrect between the caption and profiles for panels (a) and (d), Specifically, it appears that methane (solid line) is wrong in panel (a). Also, it seems that POC should be labeled as reactive, given that there is POC in deeper sediment (although see notes above). Lastly, it would be useful to have a horizontal line (or better yet shaded boxes) marking the HORM and "pre-HORM" intervals.

The HORM front is now mentioned in the figures. The figures have also been changed to color.

- Page 7665 -

Figure 7: As above for Figure 5. Also, I am uncertain what is being modeled or shown in regards to the lower SMTZ, as there seems to be no sulfate. Is the "CH4 front 2" below the base of the panels?

A new panel (panel c) has been added which clearly shows the double SMTZ. For panels d-f sulfate is no longer present in the panel and the lower SMTZ is no longer visible.

3 Reply to anonymous reviewer

This is remarkable paper. It is written with clarity, with has a strong physical and scientific foundation, it is thorough in its literature review, its results are impressive, and its analysis is insightful almost flawless. This reviewer thoroughly enjoyed reading it, and would recommend acceptance as is had it not been for a few technical issues that need either to be clarified or addressed. Because he is unsure whether the short-

BGD

8, C6148–C6167, 2012

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



comings were the results of imperfect descriptions or unsatisfactory approximations, this reviewer chose the "minor revisions" option, but it is possible that the "technical corrections" option may be more appropriate.

The issues are as described below:

(1) *The treatment of porosity: On pages 7631 and 7632, the authors state the assumptions that (a) porosity and phase velocities are invariant in time due to the assumption of steady-state compaction ..., and (b) the porosity profile is exponential with depth and time-invariant. Given the earlier assumption of steady-state compaction, it is impossible to see how the two can be compatible. Steady accretion of depositional materials and compaction indicate (by definition) a continuous decline in porosity as a function of time at any point in the profile. The only possibility of this non-happening is the employment of a moving coordinate system (which is certainly not the case here). Additionally, the issue that has not been addressed in the discussion (and which can potentially have an important impact) is that of the water level variation over time: this reader is left with the impression that the reference is the ocean floor, which is assumed to occur at a fixed depth over the period of the study. Are the authors sure that this is the case? If so, they must clarify their conviction. If not, they have to address the issue: a changing water level over time affects pressure, compaction, and, consequently porosity. Given the fact that porosity cannot be time-invariant, the phase velocities cannot be time invariant either. The authors need to provide a solid basis for their approach/approximation, and either provide convincing evidence that their approach is valid, or rerun some of the simulations with a time-variant porosity (there are several robust models of porosity change as a function of pressure, and pressure can be described as a function of time). Of course, if counter to this reviewer's impression, porosities and phase velocities are indeed time-dependent, then this needs to be better described.*

The reviewer's concern regarding the treatment of porosity is valid, and may arise due to poor word choice when describing the model. We use the term "steady-state compaction" as defined by Berner (1980). Here, he described it as

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



$\partial\phi/\partial t = 0$, where ϕ is the porosity, and t is time. This implies that under steady-state compaction the porosity profile becomes invariant in time. In this study we also apply an exponential function to describe compaction and the resulting decreasing-with-depth porosity profile. These two concepts are not mutually exclusive. Together, these concepts simply state that the porosity profile is described through an exponential function and is invariant in time.

Since the model begins with pre-Littorina sediments already deposited in the Basin (see Arkona Basin section), the model assumes that compaction has taken place in these sediments to the point of reaching a steady-state. While 35 m of muds were unlikely to have accumulated over the Holocene, the model thus assumes that pre-Holocene sediments (i.e. Pleistocene mud and glacial till) compacts to the same degree as the contemporary Holocene organic rich mud.

The second point the reviewers makes with respect to the model construct pertains to the treatment of the water depth. We have clarified this stipulating that the shoreline displacement graph in Figure 3 refers to sea-level drop with respect to contemporary sea level at Arkona Basin over the time being simulated. The reference plain remains the sediment-water interface ($z=0$). Changes in water level are only used when calculating the hydrostatic pressure and, subsequently the methane solubility concentration.

The reviewer also mentions the connection between pressure and porosity. Sediment compaction is governed by the effective stress of the sediment. Previous studies have shown that the effective stress in sediments has an increasing logarithmic relation with depth (Boudreau and Bennet, AJS, 1999). This concept has been used by several authors to independently determine level of compaction in sediments (L'Heureux and Fowler, 2000; Mogollón et al., 2009; Jourabchi et al., 2010; Mogollón et al., 2011). Ultimately these studies found that compaction is subject to the hydrostatic pressure gradient as opposed to the hydrostatic pressure itself, that is, that pressure will not affect compaction. In the particular case of Jourabchi et al. (2010), the authors demon-

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

strated that an exponential relation for porosity was a sufficiently good analogue to describe compaction.

(2) *The treatment of temperature: It is somewhat strange that the authors limit themselves to a terse statement that "temperature is modeled explicitly and modeled as a conservative species ..." (page 7635), when they describe all other conditions and properties in minute detail. This reviewer would like to suggest that readers are probably disinclined to hunt in the literature for references, and would much rather prefer a stand-alone paper that does not send them on a paper chase.*

Setting this issue aside, the treatment of temperature requires a much more explicit discussion and a much better explanation. Temperature is NOT conserved, energy (heat) is. Thus, at a minimum, the authors' statement is unfortunate. In addition: How are the temperature boundaries treated? How is the geothermal heat flux treated? What is its assumed value, and how does it change over time (if not, why not)? Do the authors perform some kind of initialization? Do the authors have some outside evidence of temperature distribution over time? If so, why do they not impose it as a (time-variable) boundary condition? How is this "explicit modeling" performed? In the revised submissions, the authors need to address these two issues, and to provide satisfactory explanations.

We agree with the reviewer that the treatment of temperature in the model was not properly addressed. We have included a more detailed description of the model treatment of temperature (the governing equation, as well as the boundary conditions). We have also rephrased the sentence concerning the implementation of temperature in the model: "temperature changes due to chemical reactions and phase changes are small enough to be ignored".

Thermal diffusion modeling indicates that the seasonal variability in bottom water temperature extends several meters in the sediment, below which temperature is constant (Dale et al., 2008; Mogollón et al., 2011). The sedimentary domain

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

of the model was 30 m. Assuming an average geothermal gradient of 24 °C/km, the temperature at the lower boundary would be 0.7 °C higher than the mean temperature at the sediment surface. This temperature difference will have a minor effect on methanogenesis rates, methane diffusion and methane gas solubility, and is therefore ignored. Rather, the temperature at the lower limit of the model is set to the Holocene temperature curve in Figure 3 since heat penetration through the upper 30 m is rapid relative to the time-scale of the simulation.

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Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



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Comment**

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