

Response to Review n.2: Volker Brüchert

November 25, 2019

General comment

“I have a lot of respect for the sophisticated details of the diagenetic reaction-transport model BRNS described in the manuscript by Puglini et al. It is a sophisticated, well-established model framework and has been used in many important publications, not the least already in the sensitivity analysis of anaerobic oxidation of methane in many different marine settings. This study takes advantage of the long developmental work that has been done previously with respect to AOM with this model. Here it is used to simulate sediment methane cycling for one of the big hotspots for potential future marine methane emissions - the East Siberian shelf sea, with its potential for thawing submarine permafrost and the potential presence of gas hydrates (although the presence of both is often contested in the literature for good reasons).”

Response: We would like to thank the reviewer for his appreciative, extremely constructive and insightful comment that not only sheds light on some critical aspects of our manuscript and helps to improve the quality of the manuscript, but also provides an opportunity to provide important clarifications and/or further detail.

Here we would like to stress that we included in the model a methane source from below (assuming different methane concentration spanning the range from 0 to the saturation concentration) which is supposed to resemble any kind underlying source. Our focus is in the upper 3 m of the sediments and we do not investigate and/or specify any explicit origin of the methane coming from below nor the model is, in such a version, sensitive to this origin. Since the area of interest is the ESAS, we hypothesize that subsea permafrost or gas hydrates may be the origin of such methane, but no results rely on this specific assumption. In fact we just wanted to stress the potential character of the non-turbulent methane emissions we found.

“The model uses the conventional setup of a network of biogeochemical reactions directly or indirectly coupled to the degradation of organic matter deposited at the sea floor. The paper is mostly not about the Siberian shelf, but is a very thorough assessment of AOM dynamics with explicit treatment of upward flow, bioenergetics controls of AOM, and a complex reaction network of biogeochemical redox reactions

as they may occur in Siberian shelf sediment”

Response: While the reviewer is absolutely right in pointing out that the results of the comprehensive sensitivity study described in the manuscript are universally valid, we would like to stress that the model setup and the sensitivity study have been specifically designed with the aim of assessing the fate of dissolved methane released from a deep source (*e.g.* dissociating hydrates or thawing subsea permafrost) in warming Siberian Shelf sediments. More specifically:

- The model is forced with a variable flux of dissolved methane potentially originating from dissociating methane hydrates and/or thawing permafrost in the deeper sediment. The methane flux is constrained by assuming lower model boundary methane concentrations ranging from 0 to a maximum concentration that is constrained by the saturation of dissolved CH₄ under pressure, temperature and salinity conditions encountered on the Siberian shelf.
- All model boundary conditions, forcings and parameters (Tables S5 and S6) are chosen to be representative of environmental conditions encountered on the Siberian shelf.
- The range of boundary conditions and parameters tested in the steady state sensitivity study are constrained based on data compiled for the Siberian shelf.

As a consequence, the study presented here does not cover the entire range of possible conditions (*e.g.* methane fluxes, active fluid flow, organic carbon concentrations etc.) encountered at the global ocean seafloor, but is representative for conditions (likely) encountered on the present and future Siberian Shelf.

“The manuscript is well written up section 3.3.1., after which it deteriorates conspicuously”

Response: We agree that the logical structure of section 3.3.1 could be improved and have carefully revised this part.

“In principle, there were two objectives: 1. BROADSCALE simulation of AOM dynamics: It does a very good job at simulating a range of broadly set environmental conditions with direct impact on the filter efficiency of anaerobic methane-oxidizing microbial consortia that use methane and sulfate. The range of the environmental conditions is set broad enough to encompass conditions that may be encountered on the East Siberian shelf. However, this part is not very novel and AOM dynamics and filter efficiency have been reviewed by Regnier et al. (2011) previously. Therefore all sections of the manuscript that relate to the simulation tests should be significantly shortened.”

Response: We strongly disagree with this comment. Regnier et al., 2011 present a comprehensive review of previously developed models that have

been applied to investigate a large employed to simulate a large set of diverse depositional environments affected by intense methane cycling, ranging from mud volcanoes and active seeps to passive sediments experiencing groundwater discharge or high organic matter inputs. The review explicitly explores how different model implementations/formulations (with increasing complexity of the biogeochemical network) perform in simulating methane-affected sediments, as well as explore simulated AOM efficiency in response to a discrete, non-specific set of environmental conditions considered in these models.

However, the analysis of AOM filter efficiency and CH_4 effluxes presented has a completely different focus and goes well beyond the analysis presented in Regnier et al., 2011. As pointed out above, the main aim of this model study is to specifically investigate the potential escape of dissolved methane released from a deep source (*e.g.* dissociating hydrates or thawing subsea permafrost) from warming Siberian Shelf sediments. It thus assesses the efficiency of the microbial AOM filter in attenuating potential dissolved permafrost/hydrate methane fluxes under a continuous and specifically chosen range of environmental conditions/scenarios (likely) encountered on the present and (idealized) future Siberian shelf using an identical model set-up and thus offering not only more robust theoretical consistency and comparability. The main focus of the presented sensitivity analysis lies on identifying environmental conditions (and thus potential areas on the Siberian Shelf) that favor non-turbulent dissolved methane fluxes across the sediment-water interface.

We further emphasized this point in the manuscript by modifying the introduction and abstract accordingly.

“2. Regional application: The second part of the manuscript is the application of the model to the East Siberian shelf. I found this part the more relevant one, given the title, but unfortunately also less well constrained due to the paucity of data used to constrain their model in face of the diversity and size of the targeted marine region. For reference, my guess is that the authors would certainly not model the whole of the North Sea or the Baltic Sea with this model, two marginal seas of similar size or even smaller than the Laptev Sea”

Response: We also disagree with this statement. One strength of a models is that it can provide the explorative means to assess dynamics at spatial/temporal scales that cannot easily be assessed by observations alone. In particular, transfer functions, simple look-up tables and/or neural networks that are derived from or trained on a large ensemble of individual model simulations over a broad range of plausible boundary conditions have been frequently and successfully used to investigate regional and even global dynamics.

For instance, Gypens et al., 2008, Dale, Nickelsen, et al., 2015, Dale, Graco, et al., 2017, Capet et al., 2016 use simple transfer functions derived from a large ensemble of 1D diagenetic model simulations to predict benthic

nutrient recycling fluxes for the coastal North Sea (Gypens et al., 2008), the Peruvian Upwelling system (Dale, Graco, et al., 2017), the entire global ocean (Bohlen et al., 2012; Dale, Nickelsen, et al., 2015) or the entire Black Sea (Capet et al., 2016). Marquardt et al., 2010 used a transfer function to estimate the global gas hydrate inventory in marine sediments. In addition, Bourgeois et al., 2017 used a generalized additive model to calculate oxygen fluxes through the sediment-water interface for the entire Arctic Ocean and Artificial Neural Networks have been used to estimate sulfate (Bowles et al., 2014) fluxes through the sediment-water interface on a global scale.

These approaches are similar to the regional assessment presented here and illustrate the power of such transfer functions. We now highlight this in the introduction.

“My specific critique relates to the following points, which to my opinion are important in controlling the biogeochemical rates and flux output of the model, but that are not or too poorly constrained in the model to substantially further our understanding of how efficient anaerobic methane oxidation is and will be in the Siberian shelf sediments. Even with the reduction of the investigated area to the Laptev Sea only, the depositional environments and geological settings are so much more variable that a simple sedimentation rate/bathymetry-based prediction of present-day organic carbon accumulation gives a starting condition for the model that is too simplifying to be acceptable.”

Response: The results of the extensive sensitivity study presented here clearly indicate the sedimentation rate and active fluid flow exert the dominant control on the escape of methane derived from thawing permafrost and/or disintegrating methane gas hydrates through the Siberian shelf sea floor across a wide range of contrasting environmental conditions encountered in this depositional environment. Results show that additional environmental conditions, such as OM content or AOM efficiency (*i.e.* k_{AOM}) play a minor or negligible role. Sedimentation rate can thus be used to predict the non-turbulent of methane escape on the Siberian Shelf.

The extensive sensitivity study presented here, thus also confirms the general approach that underlies the ensemble of studies listed in the previous response: single benthic biogeochemical characteristics, such as seafloor fluxes, redox horizons or inventories are often controlled by a limited set (1-2) of dominant factors that can then be used to robustly predict these characteristics on a regional/global scale.

“For example, the authors rely on a selected handful of Pb-210 data (there are more available in the literature for better coverage (see Bröder et al., 201; Strobl et al., 1988) for sedimentation rates”

Response: We thank the reviewer for the suggestions. Bröder et al., 2016 reports values for two sites in the East Siberian Sea and can thus unfortunately not be used to improve data coverage in the Laptev Sea. However, the reported linear sedimentation rate ($0.14 - 0.15 \text{ cm yr}^{-1}$) is not only sim-

ilar to the sedimentation rate used in our local model application (0.12 cm yr^{-1}), but would also not change flux calculations if applied (see sensitivity study). We now include the values reported by Strobl et al., 1998. They show that sedimentation rate in the Laptev sea is of the same order (0.15 cm yr^{-1})- a value that falls well in the range we explored.

“The model doesn’t consider the regionally diverse sediment types, permeabilities and rates in the Siberian Shelf Sea (see for example Dudarev et al., 2006 Oceanology; Rekant et al., 2015). The model doesn’t consider known clay/sand/sand grain size variation and their influence of carbon concentration, permeability, transport, and resulting biogeochemical rates.”

Response: We would like to stress again that the presented study does account for the regional variability of sedimentation rate: 1) in the sensitivity study considering a large range spanning almost two orders of magnitude ($0.03 - 1.5 \text{ cm yr}^{-1}$), and 2) in the regional analysis that applies a spatially variable sedimentation rate. In addition, the influence of the amount of degradable OM has also been tested in the sensitivity study and, because it is of secondary importance, is qualitatively discussed in the regional study.

It is however correct that we assume a porosity profile, which is representative for fine-grained shelf sediments. This is in agreement with Dudarev et al., 2006 (although they focus on the East Siberian Sea and not the Laptev Sea). They suggest that: “*The distribution of sediments demonstrates that they sustain fine-grained texture in the major part of the continental shelf regardless of the distance from the shore*”. Considering that the overall geomorphological characteristics of the East Siberian Sea and Laptev Sea are similar, we can assume that a 3 m sediment column with a prescribed porosity (dependent on depth) and a uniform texture and sediment type might be a decent representative for a large setting of the ESAS. We added a comment to the methods section.

“The model assumes Barents Sea depositional conditions as a good analog, however, these are unlike those of the Siberian shelf, since the Barents Sea is much deeper, has higher marine productivity, less ice cover, and much less input of terrestrial organic matter. In addition, it does not have terrestrial permafrost underneath the recent Holocene sediments. It is therefore not a particularly good analog. If the authors are interested, I can provide porewater methane, sulfate and ammonium data from this region.”

Response: We would like to thank the reviewer for this offer. We have been in contact with the reviewer for porewater methane, sulfate and ammonium data and now include an additional model test case for this Laptev Sea site. We would however also like to stress that we do not consider the Barents Sea shelf offshore Versterålen as a good analog for the ESAS. Due to the paucity of observational data from the Laptev Sea for model testing, we used this Arctic site to illustrate the performance of our model set-up in simulating biogeochemical dynamics in high-latitude shelf sediments.

“The reactive continuum approach employed here probably overestimates the reactive organic carbon amount that is available to organic carbon degradation at depth. In reality, the reactivity of the organic matter below the oxic horizons is one to two orders of magnitude lower than commonly observed in marine shelf sediments (see Figure 9, Brüchert et al., 2018). Given the very low reactivity of carbon in these sediments (See Brüchert et al., 2018; Bröder et al., 2016; Tesi et al., 2014), sulfate is likely never exhausted and methanogenesis and AOM may not even take place in these sediments at all. I am therefore not surprised at all that the authors arrive at such low regional dissolved benthic methane fluxes, seemingly at odds with the broadly published claims of extensive methane emission from the Siberian shelf.”

Response: This is a misunderstanding which we would like to clarify. First of all, we would also like to emphasize again that, according to our findings, the organic matter reactivity only exerts a secondary effect on our conclusions and therefore does not alter the overall picture of our results. In addition, we would like to stress again that the focus of the presented analysis centers on the fate of methane fluxes from thawing permafrost and/or disintegrating methane gas hydrates and not in-situ biogenically produced methane for which OM reactivity may play a more important role. The presence of a deep methane flux from thawing permafrost and/or disintegrating methane gas hydrates also ensures the presence of an AOM and the depletion of sulfates.

However, apart from this, we also disagree with the overall comment that the reactive continuum model (RCM) overestimates reactivity in these sediments. In fact, the RCM accounts for the decrease of OM reactivity with sediment depth/degradation state. Here, we test a wide range of RCM parametrizations (*i.e.* a) including those that result in a rapid decrease of OM reactivity by 1-2 orders of magnitude. Moreover the two papers cited actually support the use of a reactive-continuum model.

1. Bröder et al., 2016 show that the half-life of the organic matter deposited at two sites in the East Siberian Sea is 19 – 27 yr. These half-life are represented by our RCM parametrizations in the intermediate range. Assuming $\nu = 0.125$ the corresponding a for the two samples would be $a = 3.4 - 4.8$ yr - values that are well within the range explored in our sensitivity analysis.
2. Tesi et al., 2014 in their conclusions clearly state: “*Therefore our results suggest that TerrOC is made of several allocthonous pools each with distinct reactivity toward the oxidation (i.e., reactive continuum)*”.

We modified the method section to clarify this point and also added the two references.

“In fact, these fluxes confirm my own direct measurements of porewater methane concentrations and methane fluxes from a range of stations investigated in the summer of 2014 during the SWERUS expedition with the Swedish icebreaker Oden. If the authors are interested, I am willing to share these data with them to better constrain their model.”

Response: We are really thankful for this offer and have been in contact with the reviewer.

“The model doesn’t consider Holocene sealevel change to elaborate on the mass of sediment available for methane generation since the last glacial maximum, which is the time since reactive sedimentary organic carbon accumulation began.”

Response: This is a misunderstanding. Again, the focus of the presented paper is on the fate of methane released from subsea permafrost/gas hydrates on the present-day and future Siberian shelf. We do not intend to simulate the historical evolution of the SSPF and of related historical methane emission, but only a plausible range of current/future ones. Furthermore, our model analysis is based on the simulation of the first 3 meters of sediment and the Holocene sedimentation rates we explored ($0.03 - 1.5 \text{ cm yr}^{-1}$) indicate that the sediment layer overlying the subsea permafrost always exceeds 3 m.

“The model design relies on a sequence of thermodynamically regulated terminal electron acceptor reactions driven by fresh carbon accumulation at the top of the model domain. In reality, non-biogenic or old Pre-Holocene-produced methane transport from below (of thermogenic or Pleistocene age, i.e., terrestrial) is the key unique characteristic of the Siberian shelf with respect to methane cycling. This carbon is old and uncoupled to recent carbon accumulation. In addition, carbon accumulation varied greatly through time on the Siberian shelf. The model appears to assume continuity of recent depositional conditions back in time and space, which is most certainly incorrect.”

Response: This is a misunderstanding. In fact, the model analysis focus on this “non-biogenic or old Pre-Holocene-produced methane transport from below (of thermogenic or Pleistocene age, i.e., terrestrial)” and not on the in-situ produced biogenic methane. Because it is impossible to reconstruct depositional conditions over the Holocene for the entire region, we indeed assume broadly similar depositional conditions during the Holocene. This is an acceptable simplification, in particular because:

1. Early diagenetic rates are highest in the shallow, young sediment layers and decrease rapidly with depth. As a consequence, biogeochemical dynamics are mostly affected by recent depositional conditions. This is especially true in the light of the fast decrease in OM reactivity reported by broder 2016; Brüchert et al., 2018; Tesi et al., 2014.
2. Our comprehensive sensitivity study indicates that OM degradation and biogenic methane production in the Holocene sediment layer ex-

erts a minor control on non-turbulent methane fluxes across the sediment-water interface. Holocene fluctuations in environmental conditions will thus exert a negligible effect on our results.

We clarify this throughout the manuscript (see previous replies).

“Only the section with the transient model scenarios therefore applies to the Siberian shelf and only scenarios with an explicit upward flux of methane are relevant for investigating AOM dynamics in these sediments. However, because of the difficulties in constraining the regional distribution of seeps, flux rates cannot be reliably extrapolated and one should refrain from a regional flux estimate.”

Response: This is a misunderstanding. All steady-state simulations also apply an upward flux of methane (as outlined in the method section for details). They are thus relevant for investigating the fate of permafrost/hydrate derived methane in the Holocene sediment column and its possible escape through the sediment water interface. They also allow to derive the transfer function for possible non-turbulent methane escape that has been used to establish a regional estimate. We clarify this point throughout the manuscript (see previous replies).

Because our steady state analysis shows that AOM acts as an efficient biofilter and mostly prevents non-turbulent methane escape from the sediment, we also explored a number of plausible transient scenarios to explore if microbial dynamics could possibly create “windows of opportunity” for methane escape and assess their importance. We further clarify this in the introduction and method section. in the transient analysis we performed we actually refrained from an upscale estimate and we just explained the result of the flux out of simulated sediment column.

“My objections to the present manuscript are therefore not whether the model’s capabilities are useful to the scientific community in general, which it certainly is, but a critique of the attempt to mimic biogeochemical as well as recent and past depositional conditions on the Siberian shelf to better predict sediment methane emissions from this region.”

Response: see responses above.

“I am fully aware of the infected discussion of the relevance of the Siberian shelf sea’s role as a potentially huge methane source to the atmosphere put forward by Shakhova and co-authors. The outcome of the model simulations presented here, even in their most generous state (high advective upward flow and moderately to high sedimentation rates), would imply that the emissions proposed by Shakhova and coauthors are very hard to achieve without invoking massive gas emissions (which are not seen regionally in atmospheric measurements).”

Response: This is indeed one of the conclusions of our analysis.

“However, the inability of this 1D model to encapsulate environmental conditions

that are found in the Laptev and East Siberian Sea make it impossible to use its scaled model output to the current system or to use the model to make reliable assessments of how the shelf environment may change methane fluxes in the future. Particularly the latter requirement is key to the use of a reaction transport model such as this one in climate science. [...] The study and conclusions give the false impression that this particular model is capable, with certainty, to predict the non-gaseous methane flux emanating from this 1.5 million square kilometer large region, if one only knows the sedimentation rate and water depth. The authors may therefore consider a new title for their manuscript for the first section and resubmit it under this new title without much reference to dissolved methane emissions on the East Siberian shelf, since this is not what they can model reasonably with the data they have available. [...] Alternatively, the model simulations can be tested with actual data from the Siberian shelf, which I am willing to share. In this case, I would suggest to reduce the first part of the manuscript and focus on the application of the BRNS to the Siberian shelf sea rather than a broad treatment of the model's performance."

Response: This comment reflects a string of misunderstandings. We do not aim at quantifying, "*with certainty*" the exact evolution of present and future methane emissions from the Siberian shelf. As highlighted in the title, abstract, introduction, the presented study assesses the potential for non-turbulent methane escape (derived from deep sediment sources such as permafrost/gas hydrates) from Siberian shelf sediments. As pointed out in the results and conclusion section, it thus provides a robust, quantitative framework suitable to make first order estimates and draw conclusions with respect to present and potential future emissions, as well as methane gas emissions required to support previous estimates of Arctic Ocean methane emissions to the atmosphere. Given the urgent need to assess this potentially ticking time bomb, but the paucity of observational data, it represents a feasible and robust quantitative first step towards a better assessment of the threat methane emissions from thawing subsea permafrost/ disintegrating methane hydrates pose for our climate.

Therefore, we are convinced that the title, as well as the approach of the presented study adequately reflect its scope and do not give a false impression. However, we have adapted the abstract, introduction, method and conclusion sections to further clarify these points. In addition, we have also included a new case study for the Laptev sea site based on the data provided by the reviewer.

Specific comments

Page 8: "This is a crude overgeneralization. The authors must provide more references on the physical oceanography of the Laptev Sea and its sediment distribution and bathymetry to justify this comparison. The Norwegian setting has much higher primary productivity, is up to 8 times deeper and has substantially less ice cover over the year. If anything, the Vesterålen site shares very few similarities with the Laptev

Sea or the East Siberian Shelf Sea.”

Response: This is a misunderstanding. As pointed out in the response to general comments, we used the Hola trough sediments merely to assess the ability of the model to simulated carbon and sulfur dynamics in high latitude shelf sediments porewater profiles in a Northern shelf. No calibration of the BRNS or other following results relies on the simulations performed to reproduce the Vesterålen site, nor do we claim any similarity with the shelf areas of the East Siberian Arctic shelf. However, we do agree that our statement could be misunderstood and have now modified this section accordingly.

Page 12: “Please correct, not for methane”

Response: “Simulation results show an overall satisfactory agreement with measurements except for methane.”

Page 13:

- “It is not correct to make reference to the ESAS, since the range of the environmental conditions applied here is sufficiently broad to be applied to a wide range of shelf and slope margin settings with possible AOM. One condition worthwhile exploring and not done here is whether at low OM reactivities, the consumption of sulfate may not be completed for the time span of Holocene sediment accumulation on the ESAS (i.e., since ca 7000 years ago).”

This is a misunderstanding. As stated earlier, we investigate the fate of methane from deep sources (permafrost/hydrate) rather than in-situ produced methane (although the model also accounts for biogenic production in the Holocene sediment layer). As a consequence, we apply a range of methane fluxes from below that ensure a consumption of sulfate. With respect to the comment on the environmental conditions, we would like to repeat our response to a similar general comment here.

“While the reviewer is absolutely right in pointing out that the results of the comprehensive sensitivity study described in the manuscript are universally valid, we would like to stress that the model setup and the sensitivity study have been specifically designed with the aim of assessing the fate of dissolved methane released from a deep source (e.g. dissociating hydrates or thawing subsea permafrost) in warming Siberian Shelf sediments. More specifically:

- *The model is forced with a variable flux of dissolved methane potentially originating from dissociating methane hydrates and/or thawing permafrost in the deeper sediment. The methane flux is constrained by assuming lower model boundary methane concentrations ranging from 0 to a maximum concentration that is constrained by the saturation of dissolved CH₄ under pressure,*

temperature and salinity conditions encountered on the Siberian shelf.

- *All model boundary conditions, forcings and parameters (Tables S5 and S6) are chosen to be representative of environmental conditions encountered on the Siberian shelf.*
- *The range of boundary conditions and parameters tested in the steady state sensitivity study are constrained based on data compiled for the Siberian shelf.*

As a consequence, the study presented here does not cover the entire range of possible conditions (e.g. methane fluxes, active fluid flow, organic carbon concentrations etc.) encountered at the global ocean seafloor, but is representative for conditions (likely) encountered on the present and future Siberian Shelf.”

- “Please correct to : ’to the SWI’ The model does not provide any constraint on the SWI flux, i.e., the benthic flux itself, because here other processes play an important that are modelled here.”

Response: We are not sure which processes the reviewer refers to, but in addition to diffusion and advection, the model explicitly accounts for bioturbation and non-local transport (through bioirrigation or ice scouring). It thus provides a robust representation of transport through the SWI.

- “Referencing this study to other studies that show a range of 5 orders of magnitude in methane fluxes to justify its applicability seems odd. Please clarify how exactly each of the referenced studies supports the model findings in your simulation.”

Response: The referenced studies offer a comparison with respect to the fluxes, as well as the flux variability in response to different environmental conditions we simulated.

- “Which value was that? Not clear from the text. Apart from that, I deeply object to the use of one value to the whole of the ESAS. What is the purpose of this upscaled value? The original model value doesn’t gain any more legitimacy from upscaling and the fact that the upscaled value may be in the range of expected values neither. Please delete this section”

The maximum value we found was $27.48 \mu\text{molCH}_4 \text{ cm}^{-2} \text{ yr}^{-1}$. We added the exact value to the respective section. As pointed out in the earlier response, model results provide a robust quantitative framework to evaluate the potential for non-turbulent methane escape from the Siberian Shelf. The purpose of upscaling the maximum value to the ESAS is simply to offer an upper limit for this possible non-turbulent methane flux and show that, even if the most favorable conditions for methane escape were to be found over large shelf areas (note, this is

different from claiming that they are), non-turbulent methane fluxes would still be negligible and would not be able to support earlier estimates of methane emissions to the atmosphere.

Page 14:

- “This is an interesting conclusion. How can one reconcile the observation that methane concentrations in the methanogenic zone generally tend to increase with depth, i.e., their transport away from the zone of formation is too slow relative to the methanogenesis rate?”

Response: The Damköhler numbers are defined in such a way that the transport process considered occurs in the same region as the reaction, *i.e.* we considered the methane transport within the methanogenic zone for the evaluation of Da_{MG} and the SMTZ for the evaluation of the Da_{AOM} . Simulation results reveal that methane transport is efficient within the methanogenic zone. However, comparison with Da_{AOM} shows that methane consumption within SMTZ is slower than its transport. In other words, methane can be efficiently transported to SMTZ but it is not quickly consumed there. As a consequence, methane accumulates below the SMTZ because at the SMTZ level it is not consumed and below the SMTZ no AOM occurs.

- “This is a curious assertion for the Siberian shelf system. It is wellknown that the sediments of the Siberian shelf are not reactive enough to yield significant methane. It is instead supposed that externally introduced methane from the thawing permafrost that serves as the methane source. The current model does not take external sources into account and this is the major flaw of this paper. It is actually not suited in the current version to model the processes on the Siberian shelf.”

Response: Deep (external) sources of methane are the main focus of the presented study. See response to general comments for details on biogenic methane production, methane fluxes from permafrost/hydrates.

- “This introduction paragraph is rather wordy and doesn’t say much. Can it be shortened?”

Response: we will shorten it in the finalized version of the paper, although we value the fact that an introduction might already provide the main message of what is described in detail later.

- “Please provide a reference to the ‘traditional views’. The view proposed here is not new.”

Response: We replaced “traditional” with “intuitive”. Our findings give further evidence of the dominant role of transport processes for non-turbulent methane effluxes also in modeling scenario compatible with ESAS settings.

Page 15: “What is meant by ‘margin’?”

Response: the continental margin. We could replaced “margin” with “shelf” to avoid confusion.

Page 17: “The authors should avoid trivial sentences such as this one.”

Response: it is not necessarily trivial, since a high methanogenesis might also be expected to foster a higher oxidation process and therefore accumulation of methane is not necessarily a triviality

Page 19: “I wonder whether the reactivity of organic matter in large parts of the Siberian Shelf isn’t even lower than 100 years. More 1000 years.”

Response: we also explored the $a \geq 100$ yr. As already stated in the reply to the general comment, the reactivity of the organic matter reported in other studies (*e.g.* Bröder et al., 2016) shows that a is < 5 , not far from the value $a = 10$ yr we used for the baseline simulation. In addition, a -values > 1000 years are characteristic for deep sea sediments underlying extremely oligotrophic gyres, such as the deep South Pacific. Shelf, slope and most deep sea environments are generally characterized by $a < 1000$ years.

Page 23: “The authors are conflating to independent processes into one.”

It is not clear which processes the reviewer refers to. We guess they are, on one hand, the actual AOM and, on the other hand, the precipitation of authigenic carbonate. We do not claim or mix them up and we are aware that they are two different processes but it is well established that they are not independent, since the alkalinity produced during the AOM can drive precipitation of authigenic carbonates as reported in many site all over the globe (*e.g.* Aloisi et al., 2004; Crémière, Lepland, Chand, Sahy, Condon, et al., 2016; Crémière, Lepland, Chand, Sahy, Kirsimäe, et al., 2016; Karaca et al., 2010; Luff et al., 2005; Meister et al., 2018; Pierre et al., 2012). We are simply hinting at an indirect effect supporting our findings, aware that the two processes are however well distinct and not trivially connected.

Page 24: “These calculated active and passive fluxes are so low that they are empirically not verifiable with currently available measurement techniques.”

Response: We are aware of this limit and acknowledge it in the study. However, we would also like to point out that the exact quantity of these small fluxes is of minor importance. What is important here is that the potential for non-turbulent methane fluxes from Siberian Shelf sediments, even under the most favorable environmental conditions, is extremely limited and previous estimates of methane emissions to the atmosphere would thus require the build up of large quantities of methane gas.

Page 26: “The question is more, whether biogenic methane ever forms in these sediments, as the authors likely overestimate the reactivity of the organic matter. Altogether I think that the authors arrive at the right conclusion for the wrong reasons.”

As stated previously, we disagree with this comment. Please see reply to general comment for details.

Page 28: “From this section on the manuscript becomes distinctly less well written, more typographic errors and less succinct writing. At the same time, the discussion of transient conditions is most relevant to the Siberian shelf system. This section needs to be carefully revised and improved in its writing.”

Response: We will carefully revise and improve this section.

Page 29: “A better way of explaining the discrepancy between the two methane fluxes at steady state and the transient condition would be to show the AOM rate for the two rate laws.”

Thanks for the suggestion. We add the AOM rate profile to fig. 11.b

Page 31:

- “This is hard to understand. It should be possible to extract the instantaneous apparent k_{AOM} value throughout the simulation. Ultimately of relevance is not what the k_{AOM} is at the end of the simulation, but its time-integrated AOM rate throughout the modelled transient run.”

It is actually possible to extract the k_{AOM} at each simulated time step. However, here we wanted to explain why the final, new steady-state flux in the bioenergetic formulation is different from the simulation with the bimolecular formulation and that is the reason we focused on the final k_{AOM} , its shape and values.

- “Poor English makes this paragraph hard to understand, most importantly it is not clear how the authors arrive at their conclusion with this argument”

Response: We will carefully revise and improve this section.

- “thermodynamical”

Response: Corrected

Page 32:

- “19 years”

Response: Corrected

- “The role of sulfide was not mentioned previously. Is sulfide generally an important player for thermodynamic calculations done here?”

Sulfide influences AOM it appears in the formulation of F_T , which controls the AOM in the bioenergetic approach as shown in Eq. 11. Bicarbonate appears as well, but it is rarely a limiting factor.

Page 33:

- “The wording should be reversed. An AOM biomass accounts for an AOM filter, not the other way round”

Response: we agree but we wanted to stress that in order to have an efficient AOM filter a minimum AOM biomass is needed and this quantity has been estimated to be $> 10^{10}$ cells cm^{-3} , which is of the same order of magnitude as the value we found.

- “Overall, this is irrelevant. The supply from below is what counts for the Siberian shelf, not the in-situ production, which is negligible in almost all settings except for the Eastern East Siberian Sea and the Chukchi Sea. In addition, the statement is also irrelevant in a general sense. As the supply from below is increased, so must the proportional contribution of in-situ produced methane decrease. This is not worth mentioning.”

Response: We will edit this sentence accordingly in the final version of the paper.

- “typo here: from ... to..”

Response: Corrected

- “I am getting lost with the abbreviations”
[CH₄]₋ is the methane concentration at the bottom of the sediment column.

- “As stated this is not true and must be corrected. Never did you investigate ESAS shelf sediments in this study. Modeling scenarios were investigated, of which some conditions may apply to selected environmental setting on the ESAS. The passive/active terminology strictly applies to theoretical scenarios of system behavior.[...] Seriously, the authors have not investigated these sediments directly at all and should not make a claim to have investigate them.”

Response: This is a misunderstanding. The focus of this study is not a regional simulation of ESAS shelf sediments, but to develop a robust, quantitative framework that can be used to evaluate the potential for non-turbulent methane escape driven by thawing subsea permafrost and/or disintegrating methane gas hydrates on the warming Siberian shelf. We would again like to repeat our response to one of the general comments.

“This comment reflects a string of misunderstandings. We do not aim at quantifying, “with certainty” the exact evolution of present and future methane emissions from the Siberian shelf. As highlighted in the title, abstract, introduction, the presented study assesses the potential for non-turbulent methane escape (derived from deep sediment sources such as permafrost/gas hydrates) from Siberian shelf sediments. As pointed out in the results and conclusion section, it thus provides a robust, quantitative framework suitable to make first order estimates and draw conclusions with respect to present and potential future emissions, as well as methane gas emissions required to support previous estimates of Arctic Ocean methane emissions to the atmosphere. Given the urgent need to assess this potentially ticking

time bomb, but the paucity of observational data, it represents a feasible and robust quantitative first step towards a better assessment of the threat methane emissions from thawing subsea permafrost/disintegrating methane hydrates pose for our climate.

Therefore, we are convinced that the title, as well as the approach of the presented study adequately reflect its scope and do not give a false impression.

However, we also modified this section accordingly to avoid misunderstandings.”

- “first or first-order?”

Response: Actually both first and first-order. Modified accordingly.

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