

Response to Review n.1

November 25, 2019

General comments

General comments: The abstract is very long and contains too many information. Suggest to re-write it in a more concise way. The same comment is valid for the chapter 3.3.1 Window of opportunities, here there are interesting observations, but sometimes slightly verbose. The authors indicate that the active sediments are influenced by “deep methane source”, then at the end of the paper they define that the deep methane source is ca 3 m below the seafloor, which is not exactly very deep. Would it be possible to find another term instead of “deep”? In any case, this has to be better defined at the beginning of the manuscript

Response

We would like to thank the reviewer for the overall positive comment and suggestions. We will revise the abstract and the section 3.3.1 *Window of opportunity* for the final version of the paper.

In addition, we will also clarify the term “deep”. We used the term “deep” to refer to methane sources below the simulated sediment column (*i.e.* > 3 m) not investigating the precise origin of this methane (permafrost/hydrates/thermogenic sources/in situ production) at the base of the sediment column (which could also come from even deeper depths). But we do agree that we must refer more clearly to the base of the sediment column.

Specific comments

1. Page 2 Lines 17-18: “Under these conditions, permafrost aggraded on the shelf and was subsequently submersed when rising sea level flooded the shelf during the Holocene sea transgression (12 and 5 kyr BP)”. Reference is needed
Response: We added a reference to Romanovskii and Hubberten, 2001; Romanovskii, Hubberten, et al., 2005, for the thickness after submer-sion and Bauch et al., 2001 for the sea transgression.

2. Page 2 Line 19: explain what is “gas hydrate”

Response: a state of matter in which a low molecular weight gas (like CH_4) is trapped in a “cage” of water molecules and whose structure is thermodynamically stable under specific temperature-pressure-salinity conditions that are found either in oceanic depths or beneath the permafrost (Sloan Jr et al., 2007). We will integrate a definition in the revised version of the manuscript..

3. Page 2 Lines 29-30: “The increasing influx of warmer Atlantic water into the Arctic Ocean - the so-called Atlantification”. This term need to be explained and relevant papers need to be cited. In both “Zhang et al., 1998; Biastoch et al., 2011” the term Atlantification is not mentioned.

Response: the influence of warmer and saltier waters of Atlantic origins has been identified and brought up to the attention of the scientific community already in Biastoch et al., 2011; Carmack et al., 1995; Zhang et al., 1998, but the term “Atlantification” appears only in Polyakov et al., 2017 and Barton et al., 2018. These reference will be added in the revised version of the manuscript.

4. “Page 2 Line 2: what destabilize gas hydrate? Pressure changes or temperature increase? Or what?”

Response: both pressure and temperature change are responsible of gas hydrates destabilization as reported in paragraph 3.3 of Shakhova, Semiletov, and Chuvilin, 2019. It has been suggested that in the case of subsea permafrost associated gas hydrates, temperature plays a more important role gas hydrate destabilization (Chuvilin et al., 2018; Mako-gon et al., 2007).

5. Page 4 Line 6: which are the “changes in environmental condition” mentioned here?

Response: The transient change in lower CH_4 boundary conditions and, in case of the seasonal scenario n.2, also the change in the upper boundary conditions of SO_4^{2-} . We will clarify this point in the revised version of the manuscript.

6. Page 4 Line 12: for methane emissions and fractures, it might be useful to read a recently published paper in Biogeosciences “Yao et al., 2019”. Biogeosciences, 16, 2221-2232, 2019.

Response: Thanks for the suggestion. The recommend paper indeed supports our understanding of methane transport and biogeochemistry in fracture-affected sediments and we will add a reference to the revised version of the manuscript.

7. Page 4 Line 19: What are the “passive and active sediment”? Although there is some explanation later in the manuscript, these concepts need to be explained here, as soon as they are mentioned in the text.

Response: “Passive sediments” are sediments characterized by the absence of an advective water flow. In contrast, “active sediments” are subject to a non-zero water flow pointing upwards towards the sediment-water interface. The definition in the paper is reported at page 5, line 18-19. We will define these terms earlier in the revised version of the manuscript.

8. Page 6 Line 15: what about the anaerobic oxidation of methane?

Response: The aerobic and the anaerobic oxidation of methane have been regarded as secondary redox reaction, as they are not directly involved in the degradation of the organic matter. They are described in detail later on (page 6, line 32 and page 7).

9. Page 9 Line 10: why the authors have assumed both baseline scenarios a water depth of 30 m when the average water depth of the ESAS is ~ 45 m (data from James et al., 2016)?

Response: mainly for two reasons:

- We do not expect a large difference in the results between 30 or 45 meters, as well as if we had used 60 m. The mechanisms we identify and the sensitivity we explore is expected to be largely unaffected by such small changes in the water depth. Results indicate that one of the main controls on non-turbulent methane escape is the sedimentation rate ω . Applying the formulation of Burwicz et al., 2011, ω has basically the same value for 30 m and 45 m water depth. The only factor which is sensitive to water depth is the saturation value of methane ($[\text{CH}_4]^*$). At a water depth of 30 m, $[\text{CH}_4]^* = 5.45 \mu\text{M}$ as opposed to $\sim 10 \mu\text{M}$ at 45 m. This last value might increase even more the efficiency of the biofilter, leading in case simply to a reduction of the maximum CH_4 we identified.
- The observed increase in summer temperature (Dmitrenko et al., 2011) occurs at shallower depths (~ 10 m). We wanted to investigate even shallower shelves, as they are the ones expected to be more delicate and active from the biogeochemical point of view. For this reason we set a depth halfway between the average value of 45 m (which takes into account also deeper depths, not really important for methane emissions) and shallower shelves closer to the coast.

10. Page 10 Line 28: is the trawling in the area affecting gas hydrate stability also? Is the gas hydrate close to the seafloor? Where is the real sediment depth? Which is the thickness of the sediments that is affected by trawling? Few cm or maybe 1 meter?

Response: On the Siberian shelf, gas hydrates are often associated with subsea permafrost (the so called subsea permafrost associated gas hydrates, Ruppel et al., 2017) and are located below the subsea

permafrost. Trawling can affect sediments: from centimeters to meters to a few meters (Shakhova, Semiletov, Gustafsson, et al., 2017) and, thus, is not expected to exert a significant effect on hydrate stability. In any case, we do not simulate subsea permafrost thawing or hydrate destabilization explicitly, but rather explore the fate of plausible methane fluxes from such deep sources and therefore do not make assumptions about release mechanisms and drivers.

11. Page 17 Line 13: “rapidely”.
Response: Thanks. Typo corrected
12. Page 23 lines 26-29: Would it be possible to better explain this concept here? I found very difficult to follow the reasoning here and related gas saturation concentration with precipitation of authigenic carbonates.
Response: Thanks. We will revise this section to clarify these aspects.
13. Page 24 Line 28: Lena river and Moustakh Island in the Buor-Khaya Gulf need to be included in Figures and captions. As a general rule, all the locations that are mentioned in the main text need to be reported in location maps and relative captions.
Response: The revised version of the manuscript will include a map reporting the mentioned locations.
14. Page 26 Lines 16-17: The authors indicate that Additional physical reworking such as ice scouring or dredging, or the absence of bioirrigation, which is known to be patchy in Arctic sediments could even further reduce estimated methane efflux. I would assume that these processes might enhance the methane fluxes instead since they remobilize sediments. More elaboration is needed here.
Response: The effects of non-local mixing processes are complex. They can indeed increase fluxes by enhancing transport through the sediment. However, they can also reduce fluxes of methane (and other reduced species) by increasing the flux of oxygen and sulfate into the sediment. We will revise this section to clarify this point.
15. Page 26 Line 25: “Artic’s”.
Response: Thanks. Typo corrected
16. How does it happen that “increasing sedimentation rates occur through coastal erosion”? please clarify.
Response: Coastal erosion and the erosion of coastal ice complex provide an input of debris and sediments which are sink rapidly to the sea floor (Vonk et al., 2014). Areas close to the coast are affected by coastal erosion and will thus receive a higher input of terrigenous material.
17. Page 28 Lines 33-34: “we show that methane from deep sources (ca. 3 m) reaches the sediment water interface within 7 to 20 years.” A comment on the fact that

3 meters is considered deep has been previously reported.

Response: see comment above

18. Page 29 Line 29: wording “which is in turn is determined”.

Response: Thanks. Corrected.

19. Chapter 3.3.1 this chapter is not very well organized and it is difficult to follow.

Response: We will carefully revise this section.

20. Page 33 Lines 25-26: “On the ESAS, AOM is a transport-limited process and transport parameters thus exert an important control on the efficiency of the AOM biofilter and, thus, on methane efflux”. Please rewrite in a more clear way.

Response: Since AOM is a transport-limited process, transport processes and parameters exert a dominant control on the efficiency of the AOM biofilter and, ultimately, on the methane efflux at the SWI. We will revise the section accordingly.

21. Page 33 line27: what does “sedimentation and active fluid flow” in brackets mean respect the advective transport?

Response: We simply list the two possible types of advective transport considered.

References

- Barton, Benjamin I, Yueng-Djern Lenn, and Camille Lique (2018). “Observed Atlantification of the Barents Sea causes the Polar Front to limit the expansion of winter sea ice”. In: *Journal of Physical Oceanography* 48.8, pp. 1849–1866.
- Bauch, Henning A, Thomas Mueller-Lupp, Ekaterina Taldenkova, Robert F Spielhagen, Heidemarie Kassens, Peter M Grootes, Jörn Thiede, J Heinemeier, and VV Petryashov (2001). “Chronology of the Holocene transgression at the North Siberian margin”. In: *Global and Planetary Change* 31.1-4, pp. 125–139.
- Biastoch, Arne, Tina Treude, Lars H Rüpke, Ulf Riebesell, Christina Roth, Ewa B Burwicz, Wonsun Park, Mojib Latif, Claus W Böning, Gurvan Madec, et al. (2011). “Rising Arctic Ocean temperatures cause gas hydrate destabilization and ocean acidification”. In: *Geophysical Research Letters* 38.8.
- Burwicz, Ewa B, LH Rüpke, and Klaus Wallmann (2011). “Estimation of the global amount of submarine gas hydrates formed via microbial methane formation based on numerical reaction-transport modeling and a novel parameterization of Holocene sedimentation”. In: *Geochimica et Cosmochimica Acta* 75.16, pp. 4562–4576.
- Carmack, Eddy C, Robie W Macdonald, Ronald G Perkin, Fiona A McLaughlin, and Richard J Pearson (1995). “Evidence for warming of Atlantic water in the southern Canadian Basin of the Arctic Ocean: Results from the Larsen-93 expedition”. In: *Geophysical Research Letters* 22.9, pp. 1061–1064.
- Chuvilin, Evgeny, Boris Bukhanov, Dinara Davletshina, Sergey Grebenkin, and Vladimir Istomin (2018). “Dissociation and self-preservation of gas hydrates in permafrost”. In: *Geosciences (Switzerland)* 8.12. ISSN: 20763263. DOI: [10.3390/geosciences8120431](https://doi.org/10.3390/geosciences8120431). URL: <https://www.mdpi.com/2076-3263/8/12/431>.
- Dmitrenko, Igor A, Sergey A Kirillov, L Bruno Tremblay, Heidemarie Kassens, Oleg A Anisimov, Sergey A Lavrov, Sergey O Razumov, and Mikhail N Grigoriev (2011). “Recent changes in shelf hydrography in the Siberian Arctic: Potential for subsea permafrost instability”. In: *Journal of Geophysical Research: Oceans* 116.C10.
- Makogon, Y. F., S. A. Holditch, and T. Y. Makogon (2007). “Natural gas-hydrates - A potential energy source for the 21st Century”. In: *Journal of Petroleum Science and Engineering* 56.1-3, pp. 14–31. ISSN: 09204105. DOI: [10.1016/j.petrol.2005.10.009](https://doi.org/10.1016/j.petrol.2005.10.009). URL: <https://www.sciencedirect.com/science/article/pii/S0920410506001859>.
- Polyakov, Igor V, Andrey V Pnyushkov, Matthew B Alkire, Igor M Ashik, Till M Baumann, Eddy C Carmack, Ilona Goszczko, John Guthrie, Vladimir V Ivanov, Torsten Kanzow, et al. (2017). “Greater role for Atlantic inflows on sea-ice loss in the Eurasian Basin of the Arctic Ocean”. In: *Science* 356.6335, pp. 285–291.
- Romanovskii, Nikolai N and H-W Hubberten (2001). “Results of permafrost modelling of the lowlands and shelf of the Laptev Sea region, Russia”. In: *Permafrost and periglacial processes* 12.2, pp. 191–202.
- Romanovskii, Nikolai N, H-W Hubberten, AV Gavrilov, AA Eliseeva, and GS Tipenko (2005). “Offshore permafrost and gas hydrate stability zone on the shelf of East Siberian Seas”. In: *Geo-marine letters* 25.2-3, pp. 167–182.

- Ruppel, Carolyn D and John D Kessler (2017). “The interaction of climate change and methane hydrates”. In: *Reviews of Geophysics* 55.1, pp. 126–168.
- Shakhova, Natalia, Igor Semiletov, and Evgeny Chuvilin (2019). “Understanding the Permafrost–Hydrate System and Associated Methane Releases in the East Siberian Arctic Shelf”. In: *Geosciences* 9.6, p. 251.
- Shakhova, Natalia, Igor Semiletov, Orjan Gustafsson, Valentin Sergienko, Leopold Lobkovsky, Oleg Dudarev, Vladimir Tumskoy, Michael Grigoriev, Alexey Mazurov, Anatoly Salyuk, et al. (2017). “Current rates and mechanisms of subsea permafrost degradation in the East Siberian Arctic Shelf”. In: *Nature communications* 8, p. 15872.
- Sloan Jr, E Dendy and Carolyn Koh (2007). *Clathrate hydrates of natural gases*. CRC press.
- Vonk, Jorien E, Igor P Semiletov, Oleg V Dudarev, Timothy I Eglinton, August Andersson, Natalia Shakhova, Alexander Charkin, Birgit Heim, and Örjan Gustafsson (2014). “Preferential burial of permafrost-derived organic carbon in Siberian-Arctic shelf waters”. In: *Journal of Geophysical Research: Oceans* 119.12, pp. 8410–8421.
- Zhang, Jinlun, D Andrew Rothrock, and Michael Steele (1998). “Warming of the Arctic Ocean by a strengthened Atlantic inflow: Model results”. In: *Geophysical Research Letters* 25.10, pp. 1745–1748.