

Dear Reviewer 1,

Thank you very much for your feedback. We greatly appreciate your helpful comments and suggestions to improve our manuscript.

The modifications in regards to your comments made, are shown here accordingly to line numbers of the clean version of the revised manuscript.

With respect to your major comments made

- Results: Most of the results are direct descriptions of the figures without any further take or analysis. It is very repetitive and sometimes hard to link to the further steps you take in the Discussions. It might be useful to move some of the discussion results in the Result section.

The ice conditions of our floe were very heterogenous, e.g. different snow thickness, ice thickness, ice types/structures, ice permeability, hence we are convinced that a detailed description of all ice cores will be very helpful. Especially for readers interested in comparison of ice cores from later studies with the ice cores presented in our study. To improve the overview of the ice floe characteristics, we included the following text in section 3.1.

In L164-166: “Ridged and rafted ice can be especially relevant for the methane cycling, due to the fact that they remain more consolidated even in the summer season (in comparison to FYI) and thus allow us to investigate methane-related processes in certain layers of these ice structures”.

L125-126: “To highlight the spatial variability of the sea ice physical and biogeochemical properties across the ice floe, we describe each ice core in detail below”.

- Discussion and conclusions: what are your main findings or take-home message? At the moment it is not obvious. For example, in the abstract the last sentence states ‘We point to sea ice as a potential source of methane’. This sounds very speculative with ‘we point’ rather than ‘we find’; and with ‘a potential’. Either be more assertive about what you have found, or if you must stay speculative, then suggest more alternative theories that explain your observations.

*In the abstract the following sentence: “We point to sea ice as a potential source of methane”
In L15-16 it was changed to: “We suggest that sea ice loaded with methane acts as a source of methane for Polar Surface waters during early spring.”.*

In addition to your comment, we included the following take-home messages in the outlook/conclusion section:

- In L383-385: *“Our study provides evidence that ridged/rafted sea ice structures create environments where methane oxidation occurs during the Transpolar Drift (TPD), eventually acting as a sink for methane”.*
- In L391-392: *“For the season of early spring we propose methane release from sea ice into the meltwater layer as predominant pathway. At this time, basal melt is occurring and the top of sea ice loaded with methane is still impermeable”.*
- In L401: *“Our study suggests that the excess of methane in PSW during early spring is sea ice sourced”.*
- In L406-410: *“The relative velocities of the ice and water, the influence of stratification on methane signal retention in the surface waters, and the impact of mechanical mixing from e.g. winds and tides are important factors for the evolution of sea ice-induced methane excess in seawater underneath the ice”.*

-For example, you never mention the potential of the local sediments as sources of methane in the water column, for example nearby or on the Yermak Plateau? Why could this not be the case? Please try to better highlight your key findings throughout the manuscript (abstract, discussion and conclusions).

In section 4.2 Dissolved methane in Polar surface water (PSW) we included an explanation why we ruled out the sediment sources for the methane excess in PSW, as followed:

In L331-335:” In general, methane excess in seawater could also originate from sediments. In our case, a potential source could have been the area West of Svalbard (Sahling et al., 2014; Smith et al., 2014; Westbrook et al., 2009). However, methane released from sediments are laterally transported in the deep ocean and do not reach the surface waters (Damm et al., 2005; Graves et al., 2015; Silyakova et al., 2020). Hence, the PSW remains unaffected by methane released from sediment sources further south. Based on our data and the regional oceanographic conditions, we suggest that methane release from sea ice is a source of the observed excess in PSW”.

With respect to your minor comments made

- Some English grammatical and formulation issues, as well as missing words and typos. Please read and check carefully before resubmission.

The English grammar and formulation have been checked in the revised version of the manuscript.

With respect to your individual comments made

1.Introduction:

- L25-27: Add somewhere that you are talking about enhanced methane emissions from the ocean to the atmosphere.

In L24-25 it was changed to: “In particular, sea ice retreat may quickly induce enhanced methane (CH₄) emissions from the surface ocean into the atmosphere due to the loss of its barrier function for sea-air gas exchange (Wahlstrom and Meier, 2014)”.

- L27: ‘Because the Arctic holds large natural sources of this highly potent...’ again ,do you mean the ‘Arctic Ocean’?? or sea floor or sediments?

“Because the Arctic holds large natural sources of this highly potent”: This sentence has been removed from the introduction, we have included instead a paragraph with respect to the methane reservoir in the Siberian shelf waters:

In L29-31: “Accordingly, the methane reservoir estimate in the East Siberian and Laptev Seas, ranges from 1.6 and 5.7 Gg CH₄ in the seawater, varying with season and depending on the ice cover (Shakhova et al., 2005; McGuire et al., 2009). Hence, in these shallow shelf seas, methane released from the sediment may be entrapped in sea ice during ice formation (Damm et al., 2015)”.

- L37: ‘sea ice charged with methane’ Consider using ‘sea ice loaded with’ or another term?

It was changed to: “sea ice loaded with methane” in the revised manuscript

- L39: ‘during the last years’ please add if you can a time period here to better indicate what you mean by ‘the last years’.

In L35-36 it was changed to: “The structure of sea ice transported by the TPD has undergone substantial changes since the early 1980s, shifting from thicker multi-year ice (MYI) to thinner and more fragile first-year ice (FYI; Zamani et al., 2019; Hansen et al., 2013; Maslanik et al., 2011, 2007)”.

3.Result:

- Table 1: For clarity, could you add ‘-’ when you don’t have values for the isotopic composition? I expect that for station C9, the 3 values of isotopic composition is 3 different estimates? Can you add that information in the Table caption?

We have added ‘-’ when there are no values for the isotopic composition.

The following sentence was added to the table caption: “Brine samples were taken at stations C8, C10, C11 (one sample per station), and at C9 (three samples”).

- L122: ‘has an age of 1 to 3 years, respectively’ What do you mean by respectively there?

In L121-122 it was changed to: “Backward drift trajectories suggest that our floe originated in the Siberian Sea, while the sea ice was estimated to be 1-3 years old (Wollenburg et al., 2020)”.

- Line 196: ‘at 90-100 m depth (Fig. 6a and 7a).’ You refer to Fig 6 and 7 after Fig2, without having mentioned Fig 3-5. You will have to reorder the figures to match the order you refer to them.

Figures have been re-ordered in the revised manuscript

- Figure 7: There are 6 subplots but you only label 5 of them (a-e). The odd one out is the 3rd from the top, which I think is the ice melt estimates? Please add a subplot letter and clarify the figure caption.

The subplot letter was added, and the following information was included in the caption:

“Figure 4: Vertical distribution of several parameters in the upper 100 meters, during the entire drift 4-15 June, 2017; (a) the bars on the panel show the degree of ice melt at each station estimated from the T/S profiles following Peralta-Ferriz and Woodgate (2015). Due to the early melt season, the length scale has been omitted to emphasize that this is only used as a qualitatively guidance”.

4. Discussion:

- Fig.3: Very nice figure. We can guess most of the media but they should still be annotated: Atmosphere, ocean, sea ice, snow? In (IIa), what’s the white section with blue dots? Why the gradual change in color of the blue ocean? And do all the CH₄ annotation in the ocean indicate concentrations? If so, make it more obvious. Also add a definition of the black arrows in the caption.

“Atmosphere, ocean, sea ice, snow”, were added in the cartoon.

The figure caption was changed to:

Figure 5: Potential pathways of methane in sea ice with varying impermeable (indicated in grey) and permeable sections (in white with blue dots), i.e. winter (I) and spring (II) conditions. I (a) Relicts of the initial methane signal (source) entrapped in impermeable ice. Impermeable

intermediate sea ice layers, act as a barrier for the upward/downward transport of methane (black arrow overlaid by a blue cross). (b) Residual methane signal after methane oxidation occurred in permeable sea ice (“water pocket”), enclosed by impermeable ice layers (see Fig. 6). II (a) When basal melt starts but the top layer still is impermeable and with snow cover (white layer on top of the ice), downward brine transport initiates release of dissolved methane. Flushing events trigger methane released into the ocean. (see chapter 4.1.3). (b) Un-restricted migration of methane in permeable sea ice (black dotted arrow). Ongoing sea ice melt, when freshwater from melted sea ice is released into the water underneath, resulting on a meltwater layer, where methane remains sustained during early spring. Methane (CH₄) annotation indicates concentration. Color gradient in the ocean, reflects the increasing stratification during the seasonal evolution of the upper part of the WML (in blue) into a fresh meltwater layer (in white).

- L278-279: ‘With changes in sea ice dynamics, more of this complex ice structures may be formed, which in turn may promote changes on the methane cycling within sea ice.’: here you mean to discuss implications for the future Arctic but its not obvious. Please rephrase.

In L282-284 it was changed to: “As potential response to the expected future thinning of the sea ice, an increased number of permeable pockets formed during ice ridging may lead to favored methane oxidation therein. Under these circumstances, we suggest that the methane pathways can be modified, i.e., sea ice may be considered as a sink for methane”.

- L343-344: ‘In summary, the excess of methane in the surface water clearly point to sea-ice-sourced and early melt events as most important factors for methane release.’. This seems like too strong a statement considering the evidence you have presented.

In L352-253 it was changed to: “Hence, the methane super-saturation levels in PSW, at this time of the year, is likely to be sea ice-sourced and the ongoing ice melt process influences this excess”.

- Section 4.2.2: some attention needs to be given to the English in this section, with many language choices that are not English based. Eg: ‘the more joint journey is made...’, ‘According the drift direction, one would’

The English language has been checked in the revised manuscript

5. Outlook/conclusion

A significant part of the outlook/conclusion section has been re-written and we included additional information in the revised manuscript.

- L379: ‘We suggest that sea ice methane-released into the ocean, and in this case into the PSW, is the favored pathway in early spring.’ Do you mean anywhere? In the whole Arctic? In this region only? Please add details.’

In L390-397 it was changed to: “For the season of early spring we propose methane release from sea ice into the meltwater layer as predominant pathway. At this time, basal melt is occurring and the top of sea ice loaded with methane is still impermeable. Tracing the overall transfer of methane from sea ice into the ocean is important for understanding and quantifying the dynamic contribution of sea ice for the methane source-sink balance. It is not yet clear which process contributes the largest amount of methane release from sea ice: the brine release during freeze-up in winter or during melting in spring. Both processes need to be considered and the amount of methane must be quantified. Extended analyses and robust numerical modelling of these processes within the entire sea ice-ocean (and atmosphere) system are needed to improve our ability to predict the consequences of the methane source-sink balance modifications in the Arctic Ocean”.

- L385: ‘The final fate of the methane (excess) thereafter depends on to which extent it is diluted by additional meltwater.

In L398-403 it was changed to: “Our study suggests that the excess of methane in PSW during early spring is sea ice sourced. The degree of ice melt regulates this excess through the amount of meltwater added to the surface layer by (a) ruling dilution throughout the melting period (b) affecting the stratification and the potential for the sea-ice released methane to be retained in the meltwater layer. The meltwater layer also inhibits the sea-to-air flux from deeper levels and increasingly so during its seasonal development (i.e. freshening and warming) when it deepens through various mixing processes. Further studies should estimate the amount of methane released into the atmosphere by the sea ice-to-air flux compared to the amount released by brine rejection into the marine environment”.

-What about the dilution by ocean mixing, currents, tides etc..? You don’t mention the role of stratification here. - L394-398: The overall transfer of methane from sea ice to the ocean stays the same, whether the ice and ocean ‘travel’ together or not.

In L404-406 it was re-phrased to: The relative velocities of the ice and water, the influence of stratification on methane signal retention in the surface waters, and the impact of mechanical mixing from e.g. winds and tides are important factors for the evolution of sea ice-induced methane excess in seawater underneath the ice. Dedicated studies for these processes are needed to better understand their relative importance for this context

- L390-393: You mention warmer waters and Atlantification, Atlantification also changes the vertical ocean stratification in the region. If stratification was to increase, then methane released in surface waters could be trapped close to the surface during summer, leading to potentially increased exchanges with the atmosphere (and transfers into the atmosphere). If stratification was to decrease, methane could spread deeper into the ocean.

In L408-417 it was changed to: “Finally, as long-term consequences, we consider the effects of an increased ocean heat content leading to enhanced ice melt and, hence, more freshwater discharged into the surface layer. Within the surface layer itself, a larger amount of freshwater would lead to an increased dilution effect on the methane content. The sink capacity of the surface waters for sea ice released methane may be increased, either by dilution or by mechanical mixing processes. A fresher (and perhaps thicker) surface layer ‘cap’ than today could further inhibit the exchange of methane between the atmosphere and the subsurface ocean layers through stronger stratification/isolation relative to below waters. Thus, any methane excess in the waters below this ‘cap’ would be disconnected from the atmosphere and be subject to further mixing with surrounding waters. Especially vulnerable for such changes are the areas beyond the current inflow area in the Eurasian basin, where the effect of the “Atlantification” is expected to be enhanced (Polyakov et al., 2017). Further work is required to investigate the spatial and temporal effects of the expected increase of ice-free waters in summer to methane pathways during the melt season”.

- But changes in how far from the source, the methane is released into the ocean and atmosphere. This you don't mention here. -There is also the acceleration of sea ice drift in the Arctic which means that sea ice rich in methane that formed on the Siberian shelves, is now potentially drifting further out with the TPD towards Fram Strait before melting and changing and therefore before releasing its methane.

In L384-388 it was changed to: “A faster sea ice drift (Spreen et al., 2011) resulting from a thinning ice cover may reduce the time for methane to be oxidized within the ice, leading to changes in the methane pathways. Further research should consider rate measurements of

methane oxidation mainly in ridged/rafted ice structures to determine the long-term impact of this process. On the other hand, with an accelerated sea ice transport, methane taken up in sea ice will be transported to remote areas, and released in surface waters of regions not yet affected by methane excess”.

Figure 1: Make it clearer which part of the drift is in Region 1 and which part in Region2.

It was changed to: “In red, the stations/dates located over the Yermak Plateau (Region 1) and in black, over the Yermak Plateau eastern flanks (Region 2)”.

Figure 9: Add ‘Within ridges / rafted sea ice’ for FYI and ‘Under ridged / rafted sea ice’ for PSW on the figure. The arrow for the atmospheric background signature is not great. Could you not instead have a dot, or create another color rectangle to represent standard local atmospheric ranges?

The arrow was changed for a rectangle, and your suggestions have been included in the figure

*Once again many thanks for the careful revision of the manuscript and valuable feedback.
With best regards on behalf of all co-author.
Josefa Verdugo*