Comment on Sapart et al. 2016 "The origin of methane in the East Siberian Arctic Shelf unraveled with triple isotope analysis"

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For disclosure, I have active research projects with two of the coauthors, and have worked on past projects with some of the coauthors as well. However, I had nothing to do with the research behind or the drafting of the present Sapart et al. manuscript; the first time I saw it was when it appeared in Biogeosciences Discussions.

CH₄ emissions from the East Siberian Arctic Shelf have been the subject of intense interest since reports of very high atmospheric CH₄ in the area, and later, reports that such enhancements were being driven by CH₄-containing bubble plumes from the seafloor. This manuscript presents a new dataset of CH₄ concentrations and CH₄ isotopologue studies in the sediment beneath 3 nearshore areas of the Laptev Sea, along with similar studies of CH₄ in the water column in these areas. Additional water column measurements are also provided for a far offshore site in the central Laptev Sea, near the top of the continental slope.

So, I was very interested to read this manuscript, and I strongly feel it should be published because of its unique dataset of CH_4 and CH_4 isotopologues in this region. However, there are some issues with the manuscript that should be cleared up. The other reviewers raise many important points, and I generally agree with them. The main problem with the manuscript is that it loses sight of the main results, in my opinion. The main observations are unique and should be published! See especially my comment about lines 404-407, where the reader finally is told the biggest result. The first section of the supplement needs rewritten or removed (I would vote for removed, because it's not especially critical to the arguments in the manuscript.)

Below I bring up specfic questions:

Line 45 "primary substrate glacial water" – I am not sure what "glacial water" means in this context; normally glacial water comes from glaciers... I think the authors mean water that has been frozen in to the subsea permafrost since formation, but I'm not sure. See also line 255.

Line 51-53—I don't see anything in this manuscript that says that the sediment CH₄ studied in this manuscript rapidly migrates through the water column. Bubbles were not trapped and analyzed. The last sentence of the abstract should be removed.

Line 66-70: "The four key mechanisms controlling the release of Pleistocene carbon from thawing subsea permafrost are gas hydrate degradation, thermokarst development, the deepening of the permafrost active layer and coastal erosion (e.g. Shakhova et al., 2005, 2009, 2010a,b, O'Connor et al., 2010, James et al., 2016)."

Several things wrong with this statement, it is talking about subsea permafrost but gives examples that only apply to land! (1) I have never heard of active layer deepening in subsea permafrost. This implies an annual freeze-thaw cycle, as happens to permafrost regions on land, not at sea. (2) Similarly, I'm not sure what undersea thermokarst is—thermokarst landscapes form due to seasonal cycling. Are you saying there are annual freeze-thaw cycles in subsea permafrost? (3) Coastal erosion does not, by definition, release carbon from subsea permafrost—it releases carbon from the eroding coast line. The entire sentence should be removed or rewritten.

Line 100-102: "Below this gas hydrate stability zone, CH4 occurs as free gas and can be advected towards the surface through faults in the sediment."

Why would this free gas not be incorporated into hydrates as it passes through the stability zone? Or are you suggesting that CH_4 released BELOW the gas hydrate stability zone migrates upwards so fast through the sediment that it is never trapped as hydrates?

Line 120: "vigorous bubbling" is undefined with no sense of scale.

Line 184: "expanded into a smaller flask for storage". Impossible to expand something into a smaller volume.

Line 191: The reader has no clue as to the logic behind the four core identifiers: " ID-11, IID-13, IIID-13, VD-13". To the reader, these are just random numbers and letters, and they are similar enough to be confusing. It would be far less confusing if they were simply designated background, 1,2,3 (or 1,2,3,4) in this manuscript. OR, if the core names themeselves are somehow significant, or correspond to information in other papers, that should be explained.

Line 199-201: The cores are shown in supplement Figure S1 (I think this figure should be in the main text). But in Figure S1, the white and black bars beside the cores show frozen/unfrozen. But here, it says that "IID-13, IIID-13 and VD-13" are partly frozen, yet IIID-13 is shown as completely thawed in figure S1.

Line 215: Another good reason to move figure S1 to the main text!

Line 255: Here, old water frozen in the permafrost is called "meteoritic"—so again, why is it called "glacial water" on line 45?

Line 282: " strong evidences that CH4 from old reservoirs (Pleistocene age or older) is being released there." -- or that the CH_4 is being formed from old carbon being released from reservoirs.

Line 296-297: This seems like a slight misunderstanding of the Overduin et al paper; in that core, almost all the loss of CH_4 occurred at the thaw front, not near the sediment surface / seawater interface. Compare Figure 4 in the Overduin et al paper with your Figure 2. I see no sharp cutoff in CH_4 values at the thaw front in your paper as Overduin et al report. (However, the use of a log plot in Figure 2 makes it somewhat hard to see.) Also, label the thaw front in Figure 2 for each core.

Line 340-345: " In the ESAS, the pycnocline is very shallow and a very low primary production is expected because of darkness and ice cover in the winter and because of the little available sunlight in the summer due to the high solar zenith angles and the very turbid waters (light penetrates only down to 40cm)"

The statements about light penetration depth are NOT TRUE. As written, this is about the entire ESAS. I suppose that close to shore, turbid water can occur (and can be seen from space), but farther from shore, water is not so turbid (again, can be seen from space). 40 cm light penetration depth is extremely shallow. Yes, water surfaces are more reflective at shallow light incidence angles, but there is a lot of sunlight in the summer in the Arctic! Photos have been published showing blue waters around islands in the study area in the summer. I don't know if there is in situ production of CH_4 in the water column or not, but I am 100% certain that light penetrates far deeper than 40 cm in waters of the ESAS.

Also, the authors have previously claimed (in this journal!) that the ESAS waters have high productivity! dx.doi.org/10.5194/bg-8-1745-2011

Line 385-387: "This gas is formed continuously from old substrates at depth and/or has been stored as gas hydrate and/or gas pockets in or below the subsea permafrost."

This sounds like you are100% ruling out biogenic CH₄ production in the near-seafloor sediment, where such production might happen utilizing recently deposited carbon sources mobilized from terrestrial and coastal erosion sources? Interesting that you rule that out. Really?

Line 393-394: "No quantitative estimate of this CH4 source is to date possible," Some of the papers you cite give quantitative estimates. So do some papers you don't cite. Maybe you mean something else here?

Line 404-407: " Our results show that thawing subsea permafrost emits large amounts of CH4 that is depleted in heavy isotopes and that such emissions cannot be easily distinguished from Arctic wetland emissions when looking only at stable isotope data. "

I believe this is the most important result of the study, and this potential caveat about isotopic studies in this region should also be mentioned in the abstract, and earlier in the text.

Other items:

I do not understand why we are presented Figure 5, which shows sulfate, C_{org}, chloride, and Si for only the background core. Why not for all 4 of the cores discussed here? Or, better still, because the dataset presented here is unique and will be of interest to many, I **strongly encourage the authors to make the all data shown in Figure 2 available with the manuscript**, perhaps as a supplement.

I find the title a bit curious. Saying "unraveled" suggests that the mystery has been solved; I would say it has not been (but that is okay!!). To me, there appears to be vast areas of the ESAS which have not been sampled yet for sediment CH₄. Hence, the title seems--premature. Unless the authors mean to imply that our understanding is being unraveled?

Supplementary Material

Line 5-9: Doesn't make much sense as written. Perhaps the authors mean something like: " Although thawing is the most obvious factor affecting the permeability of permafrost to gases, there are other factors to consider, which we discuss below"

Line 16: "the content of unfrozen water"—should be "the fraction of unfrozen water"

Line 22-23: "as it has been demonstrated"—should be "as has been demonstrated"

Line 27-31: Doesn't make sense as written. Groundwater and porewater are not the same but are apparently used interchangably here.

Suggest something like: "The salinity of this cryogenic porewater usually ranges between 10 and 300psu. Freezing-point depression is also due to the dissolved-solids content of this cryogenic porewater (Gilichinsky et al., 2007). The high salinity and solids content is due to inclusion of brines from the freezing of marine sediments."

Lne 32-34: Doesn't make sense as written. Suggest something like: "These water layers are usually connected to each other, building up a multi-level transport system which allows gases and geofluids to migrate through subsea permafrost and potentially be released to the water column, possibly via taliks."

Lines 35-38: redundant.

Line 35: Biggar et al study is not relevant here. It's about (as the title gives away) spilled fuels migrating downwards.

Line 40, Section 1.2.

This seems to be about terrestrial permafrost; but this section is headed "Factors affecting gas transport in subsea permafrost".

Line 44: "alterations of compression" doesn't make much sense. Maybe something like " they affect frozen soils and sediments by alternately compressing and stretching them during freeze-thaw cycles."

Line 53: Section 1.3.

This entire section is messy and difficult to read. It is also about processes happening far below the study zone of this manuscript. In my opinion, it can be removed without any loss to the manuscript.

Line 140-141: The problem with explaining the 14C-hot samples is that they are hottest at depth, right? Why would anthropogenic contamination not be greater at the top of the sediment, instead of under 30 m of sediment? That is a mystery. Seems like some comment should be made about this (at least to acknowledge the mystery.)

Line 155 "was abnormal" should be "were abnormal".

Figure S1: Should be part of main text. Label which core is the background core.