Interactive comment on "The origin of methane in the East Siberian Arctic Shelf unraveled with triple isotope analysis" by Célia J. Sapart et al.

Anonymous Referee #1

General comments

It was a challenge for Sapart with co-authors to satisfy entirely all the reviewers (including online open comments), but they rewrote the manuscript extensively and remade some figures to address successfully all questions raised by reviewers in their reviews. The revised manuscript (ms) by Sapart et al. presents valuable information on the origin of methane in the East Siberian Arctic Shelf (ESAS), long been discussed based on assumptions and speculations. I like that this data set represents multi-year results of triple-isotope analysis of water and sediment. To my knowledge, this is first presentation of such a kind collected in the marine Arctic. I made my search and found that up to date only one paper described an application of the triple-isotope analyses addressed to the marine methane origin (Kessler, J.D. et al., 2008. A survey of methane isotope abundance (C-14, C-13, H-2) from five nearshore marine basins that reveals unusual radiocarbon levels in subsurface waters. J. Geophys. Res., Oceans113. http://dx.doi.org/10.1029/2008JC004822.). This fact itself makes this ms novel as I already mentioned in my previous review. .

Specific comments

Now I am satisfied with explanations presented by the authors regarding my key questions:

- <u>The possible contribution from thermogenic sources</u>. I agree with authors that their tripleisotope dataset does not allow to totally exclude the presence of thermogenic methane in the ESAS sediment in another locations. They clearly state that "at the sediment sampling locations, the methane present in the sediment porewater is clearly of biogenic origin and no thermogenic signatures have been observed there". I think that that result combined with the literature data (e.g. Cramer and Franke, 2005; Bussmann et al., 2013) might indicate on high heterogeneity of sources in the vast ESAS region.
- 2) The possible contribution of super-modern radiocarbon in methane by anthropogenic sources. Authors demonstrated that the largest enrichment in 14C is observed at about 30m depth in the seabed suggesting that sea water cannot be the cause of this enrichment. I also agree that the most likely hypothesis to explain this highly enriched 14C values is that nuclear wastes have been deposited somewhere in the permafrost (likely inland) and that leakages from this area are contaminating the groundwater aquifer and therefore lateral underground transport may transfer organic matter highly enriched in 14C to the shelf environment including subsea permafrost. From my knowledge, I would add (no need to include in this ms) that one from sources of that enrichment could be numerous underground nuclei bombing tests performed in the northern Yakutia in the end of Soviet epoch.

And again, as I wrote in my first review: "Many other questions could be raised, but I realize that this manuscript is based on multi-year work in the harsh Arctic environment. It is clear to me, that one paper, even incorporating that extensive data set, cannot answer all scientific questions regarding the complex, and previously insufficiently studied, Arctic marine methane cycle. I appreciate that the authors have been accumulating data for a long period trying to cover as much aspects of this novel topic as possible. I also understand limitations possessed by current state of isotope biogeochemistry, which make it difficult to interpret isotope data collected in actual environmental conditions where methane of different origin, age, from different sources could be contributing differently in different areas – it differs so much from all idealized conceptions used to interpret the data". I suggest that these questions would be addressed in further work on this topic and the current ms would be taken as a baseline, relative to which results of further investigations in this area could be evaluated.

Bottomline. By my opinion it is very complicated to corroborate their explanations by independent analyses. Therefore, a special field campaign to cover a larger area (terrestrial and marine) and aiming to

extract much larger volume of water and sediments would have been required to obtain such data, but authors have either insufficient funding nor the authorization for such a deployment. Note, that getting a set of permissions to work in the Russian Exclusive Economic Zone (EEZ) needs a lot of long-term efforts and a good fortune. I admire authors' persistence exhibited in order to obtain such unique results in the Russian EEZ, including new data from the long sediment cores recovered from boreholes drilled by author's team.

I believe this new data would be of great interest to scientists working in different disciplines and areas of the Arctic: to geologists, biogeochemists, oceanographers, atmospheric scientists, climatologists and climate scientists. At this point, and to stimulate further development of this novel, original, and complex research, I recommend to make the results presented in this paper available to the scientific community worldwide and to publish this paper as is.