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Biogeosciences Discussions

Interactive comment on "Quantity and distribution of methane entrapped in sediments of calcareous, Alpine glacier forefields" *by* Biqing Zhu et al.

Anonymous Referee #2

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GENERAL COMMENTS: The topic of the reviewed manuscript (MS) is the occurrence spatial distribution and estimated total mass of methane (CH4) entrapped in calcareous sediment and bedrock in glacier forefields in the Swiss Alps. The topic is both novel and relevant for the improved understanding of terrestrial CH4 reservoirs and their potential source of emission to the atmposhere. The current study takes of where Zhu et al (2018) ends, with a clearly formulated aim (ll 87-89): "To extend the work of Zhu er at (2018) to other calcareous glacier forefields located in different regions of the Swiss Alps, and to assess the distribution of entrapped CH4 contents within and compare total mass of entrapped CH4 between all sampled glacier forefields". The study is well designed with clear descriptions of the work that has been done. However, since the study can be viewed as almost a part 2 of a larger study of entrapped CH4 in

calcareous sediment in Alpine catchments, with the Xhu et al (2018) paper as part 1, it could benefit from reflecting this more clearly. In particular, I would recommend that the manuscript is abbreviated significantly, to sharpen the focus, novelty and importance of the extended study, making it as short and concise as possible using referencing to the Zhu et al (2018) paper when it comes to more general background information. Finally, I recommend that the use of sediment age and landform as explanatory variables for the potential amount of entrapped CH4 present in the sediment is revised according to the specific comments.

In summary, I recommend publication of the study following a careful revision of the manuscript.

SPECIFIC COMMENTS: Ll 30-69: The two opening paragraphs are almost identical to the introduction of Zhu et al (2018) and does not mention subglacial/ glacial CH4. These two paragraphs could be shortening into a few referenced sentences with reference to Zhu et al (2018), in order for the MS to more quickly get to the essence of this study (from line 60 and onwards).

l.125. In this section the three steps of the fieldwork of stage I is described. However, the actual testing of the effect of sediment depth, sediment exposure age and landform on entrapped CH4 as well as total mass estimation comes after the fieldwork as it is based on data analysis. The text describing this should therefore be placed at a more appropriate stage in the MS.

l. 129. Is there a difference between a randomized design and a completely randomized design? If yes, please explain. If no, remove completely.

l. 152 What is the uncertainty on depth estimated based of the electrical resistivity tomography (ERT) method, and how does this uncertainty propagate into total CH4 mass best estimates? Section 2.2.1 in general. Can you absolutely rule out that no CH4 is chemically produced during the acid dissolution of the carbonate rock (by e.g. cold temperature version of similar processes as the high temperature

conversion of CO2 into CH4 using reduced metals as catalyst as decribed in e.g. https://onlinelibrary.wiley.com/doi/full/10.1002/cssc.200900152)

L 178. Good example of efficient referencing to previous literature on the same topic, saving space in this MS.

L. 179. Why where particles larger than 20 mm excluded?

L. 189. Maybe expand a little on how the initial tests were conducted in order to reach your methodological conclusion

L. 204. The number of samples used should be more clearly stated. Avoid using “About five samples. . .”

L.232. Title of 2.3.2 should include a description of what is being estimated, e.g. “Estimation of XXXX for five glacier forefields”.

L. 237 and onwards. Natural chemical dissolution of carbonate rock from carbonic acid is an important weathering process over geological timescales and is also likely to be a process of relevance in the glacial forelands in which the study is performed. With this time perspective in mind, the reason for choosing the time difference between 1850 (asserted latest glacial maximum) and 2018 (same but minimum) as the boundary conditions for the CH4 upscaling appear somewhat arbitrary. I understand of course that it is an operational boundary condition for identifying the area of the current glacial forelands. But the temporal relevance of the phenomena that you investigate, where the natural chemical weathering rates are in fact the rate limiting factor for potential CH4 emissions needs to be justified on a bigger timescale. Especially the implied relationship the last glacial maximum could serve as a time zero for CH4 release (see also line 326) and that the current climatic development should in fact increase emission to the atmosphere, which may very well not be the case, if the overall rate limiting factor is not glacial coverage, but rather the kinetics of natural carbonate weathering, which can take place both in a warm-based subglacial environment as well as in the current

pro-glacial settings.

L. 273. Your conclusion that the gas must be of thermogenic origin and has not been altered by physical/chemical weathering appear correct. However, this particular conclusion supports the view that sediment age (i.e. time since last exposure) could be an irrelevant measure on the time scale that you operate, which also your statistical analysis show (l. 283).

L. 283. The term Landform is a less exact term for the variable than e.g. mineralogy of the sediment, parent material for the sediment or similar. As stated in the MS in seems that you argue that the landform itself has a significant effect on the potential CH4 content, while I believe you mean that significant difference in entrapped CH4 content is observed between different landforms. The sediment in the different landforms (floodplain, terrase, sand hill, etc) could potentially originate in contrasting parent material, and the variation in entrapped CH4 is more likely an effect of this, rather than the landform itself, the time of deposition or the time since the most recent exposure to the atmosphere after year 1850. I recommend that both this section and the MS in general is revised to reflect this relationship.

L.302 Similar to the comment above, the distance between landform and its relationship to entrapped CH4 is more likely to be a proxy of the parent material of the sediment and deposition history than the time elapsed since the areas were covered by glaciers, i.e. glacier extend and sediment age as defined could be irrelevant properties for explaining the inferred amount of entrapped CH4.

L. 319 Please use a quantative terms, rather than the qualitative term “little”.

L.326 as stated in the comment above, more arguments should be provided to back up the assumption that maximum glaciated area in year approx. 1850 is the original value as stated here. It seems that, the original value could more correctly be described as the start value for your estimate of the exposed proglacial area following the most recent major glacial retreat, which could or could not have a direct influence

of the amount of CH4 stored in the sediment. However, the relevant age of the sediments CH4 content (i.e. the time the CH4 was trapped in the rock which by weathering became sediment) is not related to the point in time in which it most recently was exposed to the atmosphere (i.e. not covered by ice), nor to the point in time where the sediment was deposited in the current landform. It is highly likely that the sediment that you sample here and now, on several other points in tie have been exposed to the atmosphere without either incorporating more CH4 or releasing parts of the currently entrapped CH4.

L.329. Consider the wording in the sentence: " From these numbers, the total mass of sediment-entrapped CH4 in all Swiss glacier forefields derived from calcareous bedrock was computed. . .." I recommend that this sentence should be revised to more appropriately reflect what the study has done, namely to give a best estimate of the entrapped CH4 in an area of the glacier forefields corresponding to the area extending from the current position of the glaciers to their reconstructed position during the most recent glacial maximum, which is different from "all Swiss glacier forefields". The findings and associated increase in scientific understanding is sufficiently strong, novel and interesting in itself, and I see no reason for trying to upscale the potential amount of CH4, which at the current level of understanding will be very uncertain.

L. 376 Little variation in entrapped CH4 across sediment depth and exposure age, indicate that the CH4 concentration is not dependent on recent transformations or release, but an inherent property reflecting the CH4 content of the parent material (as also indicated in L. 383). Again, sediment age as defined does not seem to be a very relevant explanatory variable.

L.386/387. What "major alterations" do you suggest that the sediment has undergone during and after the erosion of the parent materiel? Usually, physical erosion of bedrock primarily reduces the grain size of the material in question without any further alterations to the matrix of the grain (unless the material undergoes diagenesis). I believe your observations point towards the opposite, namely that the sediment has

not undergone any significant alterations with respect to entrapped CH4 and that this property is indeed one of the key take-home messages of your story, i.e. that large quantities of entrapped CH4 is present, but not very likely to be quickly mobilized by natural weathering with following release to the atmosphere. The importance of this is of course linked to recent discoveries of subglacial CH4 emissions (as included in your references), in which the sediment entrapped CH4 is likely not a major contributor, unless there is significant subglacial dissolution of calcareous material with entrapped CH4.

L- 404/405 and 413/414. Yes, differences in CH4 content of the parent rock is likely the main explanation for the observed variability. An improved mineralogical investigation of the sediment in the various landforms would be able to test whether the sediment in the floodplain is significantly different that the other two landform, thereby providing a possible explanation for the observed differences in CH4 content.

L 426/427 What is the relevance of comparing a large, entrapped, immobilized and thermogenic CH4 volume in sediment with no proven interaction with the atmosphere to a mobile, biogenic CH4 pool in lakes, wetland and wild animals? These are two completely different carbon cycles, with very contrasting element cycling times.

L 433 - 441. Why is it important to narrow down the uncertainty of how much CH4 is indeed present in the entire area going beyond your study area, if the CH4 is not mobile?

L441 – L 453. CH4 emission release rates by chemical weathering is likely to be orders of magnitude lower than reported rates of microbial CH4 oxidation in soil and sediment. The described scenario is quite hypothetical and non-documented. To strengthen the scope of the study and highlight its importance, I recommend to remove this last section of the MS dealing with microbial oxidation and exchange with the atmosphere, as this is most likely not happening at a rate with any significance for biological CH4 turnover.

L. 455-468. Very good summary of the presented work and conclusions.

L. 469-474 Somewhat speculative when the data suggest the opposite, i.e. that CH4 is very stable within sediment and not released due to weathering at any significant rate (no significant difference with sediment depth+ entrapped CH4 in sediment reflect that of parent material). I suggest removing this part of the conclusion to make your story more focused, and not end on a speculatory note, when you in fact have quite strong and novel data.

Figure 2: Good idea to show sample point and profiles on a map. However As mentioned above, the concept of sediment age as an explanatory variable does not seem justified.

Figure 3: The difference between minimum and maximum glacial extend could be irrelevant as an explanatory variable.

Figure 7: More info on mineralogy and parent material would be useful to better characterize and understand the shown differences in entrapped CH4. The absolute unit "Mass of entrapped CH4 (t)" is very dependent on your upscaling and its associated uncertainty. I suggest to revise figure 7a, the show entrapped CH4 in relative terms (could be g CH4 per ton sediment or similar) to better the variation span in entrapped CH4 per sediment type (i.e. what you call landform).

Table 2: Sediment porosity: It is not clear if the the parameter "porosity" indicated intra-grain porosity (i.e. amount of pore volume within the sediment) or intra-grain porosity (i.e. amount of pore volume between grains, which must be assumed to be occupied by atmospheric air at approximately 1.9 ppm CH4). Please clarify.

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