Response to referee comments of manuscript "Porewater $\delta^{13}C_{DOC}$ Indicates Variable Extent Of Degradation In Different Talik Layers Of Coastal Alaskan Thermokarst Lakes" by Ove H. Meisel et al.

We thank referee #1 for the comments and the constructive discussion of this manuscript. In the following we reply to your remarks. (Any line number references (L...) are based on the originally uploaded manuscript. Our responses to the comments are written in green color and italic letters. <u>Newly written text that will be added to the revised</u> <u>manuscript is underlined</u>.)

Referee Comment #1 (Received and published: 4 January 2021)

This manuscript reads extremely well. The design of the study is described well, the methods are solid, the presentation of the data is very clear. The main finding is that the top sediment layer of thermokarst lakes has more active C cycling than the bottom layer - which stems from a different geological period and has mainly been formed as a marine deposit, and probably prior to the onset of permafrost formation. The results that are discussed most extensively are the carbon isotope data, especially delta-13-C. The discussion section is quite speculative - opening for mechanisms that drive the isotope signal both up and down. It is finally concluded that the relatively large variation of the delta-13-C signal in the top layer is likely to be related to the increased DOM degradation rates. *Dear referee #1, thank you for your positive remarks and constructive comments.*

I would suggest an equally plausible explanation, i.e. that the top layer is more heterogeneous than the bottom layer, because the C inputs here stem from 'old' (permafrost eroded soil) and 'young' (tundra vegetation) sources (how about sphagnum in the ponds? not mentioned - but the ponds are shallow and could have a thriving sphagnum community). The weakness in the data collection is that the isotopic characterization has not been matched with an assessment of DOM degradability. That could have been done with a simple dark incubation, and would have given a measure of which DOM (talik or lake sediment DOC) was most accessible to microbes. It is likely, of course, that the microbial community in the top is a lot more active (and has access to more nutrients) than in the talik. So some additional nutrient measurements (CN ratios of DOM can also give an indication of DOM nutrient depletion and how far it might be degraded) would have been helpful. So, it seems a bit as though the isotopic data (except for the 14C) actually doesn't help a lot with understanding C cycling in these lakes. I can agree with the conclusion - but not because of the evidence suggested by the 13C results. You also write yourself that the variation in DOC might be source-driven (1 505), but you seem to leave that line of reasoning later in the discussion. So while I admire the manuscript for its clarity and its great explanations of thermokarst formation, I think that the interpretation of the data has some lose ends.

We agree with the above stated main point of criticism that degradation-related data are missing to further back up the observations and conclusions made from the δ^{13} C-based data regarding the different levels of soil degradation in the individual talik layers. We would still like to argue that the conclusions based on the presented δ^{13} C data were carefully worked out and worded openly to leave room for discussion. But we agree that it is essential to back up the conclusions made in this manuscript with degradation-specific data in future studies. We will add the following section in L545 of the conclusions where we will stress the need for degradation-related data and give examples of possible analysis that would be beneficial.

<u>''In future studies this finding needs to be further verified by additional analysis, for example of C:N ratios, dark</u> <u>incubations or humification measurements of the SOM to back up the conclusions based on the $\delta^{13}C_{DOC}$ data and to rule out that the observed trends in carbon isotopes are not solely driven by the sources of sediment but degradation processes.''</u>

Nevertheless, we think that the findings are presented with a reasonable level of assumptions. We also see this manuscript as an important gap filler in terms of carbon-related data from these specific Arctic environments where the amount of available data is still scarce.

In L496-499 we discuss the possibility of a mainly source-driven $\delta^{13}C_{SOC}$ signal in the lake sediment and deeper talik layers as the input of organic material into the lakes is mainly of terrestrial origin. We point this out to emphasize that $\delta^{13}C_{SOC}$ or $\delta^{13}C_{DOC}$ data alone would not be sufficient to draw conclusions with regard to their degree of OC degradation. But with both data sets in combination the observed changes from $\delta^{13}C_{SOC}$ to $\delta^{13}C_{DOC}$ become interesting indicators of possible degradation activity as they are less likely to be solely source-driven and more likely to display in-situ sediment processes. Please see the above-mentioned sentence we will add to the conclusions section in L545 where we also point out that further analyses are needed to further confirm the results also with regard to the possibility of sediment source-driven isotope signals.

Regarding the presence of Sphagnum in the ponds please see our reply below to comment number 2.

1. I miss a hypothesis (198 would be a good place).

Thank you for pointing this out. Yes, we agree that a clearly stated hypothesis is missing and that L98 is a good place to insert one. We will add the following sentence to L98 to make the intentions of the manuscript clearer to the reader: <u>''We hypothesize that the level of soil degradation is likely more intense towards the top of the talik in the</u> <u>'lake sediment' and declines with sediment depth towards the 'deeper talik' layers.''</u>

2. please add some information about the presence of vegetation / sphagnum in the lakes (minor issue, but good to know).

For this comment, we would like to point to the below listed parts of the manuscript where the presence of identifiable (where possible) and unidentifiable plant material was discussed in the results section:

- L260 (''...clusters of unidentified plant remains...')
- L302-305 ("The sediment of Unit C is for a large part made up by plant detritus of terrestrial origin, mainly in the form of unidentified plant remains but also of roots, small leaves and stems among which brown mosses, Sphagnum and Betula nana were identified. ")
- L311-313 (''...Unit C is classified as a homogenous 'lake sediment' that contains large amounts of terrestrial plant detritus...')

Unfortunately, the majority of plant remains were degraded to such a degree that the identification was not possible anymore under a microscope. Therefore, a more in-depth discussion and quantification regarding the presence of different vegetation types found within the talk deposits was not feasible.

We agree however, that a short section mentioning the different vegetation types growing in the surrounding landscape and ponds additional to the identified plant remains in the sediment should be added to the manuscript. We will add the following sentence in L130 to the field site section of the results chapter:

<u>''The lakes are surrounded by a marshy tundra landscape with shallow ponds and vegetation consisting of tundra</u> grasses, brown mosses, Sphagnum and occasional Betula nana shrubs.''

3. a more extended explanation of the normalizations should be added to the methods.

Agreed. The explanation for the normalization of DOC concentrations and SOM amounts to DOC_{norm} and SOM_{norm} , respectively, will be explained in more detail for a better understanding of the applied methods. Please see the new text below which will replace the short description in L341-344 of the manuscript :

"The DOC_{norm} data displayed in Fig. 5a are normalized DOC concentrations converted to their total carbon mass in [mg] taking into account the known sediment volume, density and porewater content. The normalization of DOC data points at certain core depths is based on the fixed volume (V_{core}) of 1 cm (h) thick cross sections of the sediment core tubes (d = 9 cm) with a volume of 63.6 cm³ ($V_{core} = \pi^*(d/2)^{2*}h$). The total sediment density ($\rho_{sediment total}$) of each core section is known from the gamma-ray density measurements. Together with V_{core} the total sediment mass including the pore water ($m_{sediment total}$) can be determined for each core depth ($m_{sediment total}=V_{core}*\rho_{sediment total}$). The wet and dry weights of sediment samples were also measured, thus the relative amount of porewater [wt%] in relation to the total sediment mass (Fig. 2-3) at a certain core depth is also known. Together with $m_{sediment total}$, the total porewater mass ($m_{porewater}$) was calculated ($m_{porewater}=m_{sediment total}*porewater$ amount [wt%]). With $m_{porewater}$ in turn the total porewater volume ($V_{porewater}$) present at a certain sampling depth can be calculated together with the known density of water ($V_{porewater}=m_{porewater}/\rho_{water}$). In a final step $V_{porewater}$ and DOC concentration are used to determine the total mass of DOC present at a certain core depth for that fixed 63.3 cm³ volume (V_{core}) of a 1 cm thick core cross <u>section ($DOC_{norm}[mg] = DOC[mg/l] * V_{porewater}[l]$)</u>. Normalized DOC_{norm} values [in mg] display the total amount of DOC present in each individual layer better than relative DOC concentrations [mg/l]. ''

4. Figure 5a suggests to me that an equal amount of SOM delivers more DOC in the top than the bottom (1485), which a different interpretation than what you have. Please clarify.

In L484-485 we are exclusively talking about the trends in DOC dependence on SOM availability which is in fact very similar and comparable in both layers of lake sediment and the deeper talik. In L476-482 we discuss that the total amount of DOC formed is, as you pointed out as well, in fact higher in the lake sediment in comparison to the deeper talik layer based on Fig. 5a. But we see that the wording in L484-485 can appear ambiguous to the reader and will rephrase it in a clearer way in the revised manuscript:

''In Fig. 5a we also show that the formation of porewater DOC is directly linked to the availability of SOM/SOC in the surrounding sediment, as yields of DOC (as DOC_{norm}) increase with increases in SOM_{norm} content, although the total amount of DOC yield is significantly higher in the 'lake sediment'.''

6. Fig 5b (should you turn around the axes? DOC on the x-axis? more intuitive to me), here it looks like the DOC concentration drives the isotope signal and that the isotope signal in both layers is essentially the same, it's just driven by differences in DOC level. This is also shown by figure 6b (LOI and DOC were somewhat interchangeable, weren't they?) for 5a, what is the relationship if you plot it against normalized DOM?

Agreed, we will exchange the horizontal and vertical axes in Fig. 5b of the revised manuscript.

Yes, in Fig. 5b the isotope signals of the lake sediment and deeper talik appear to follow the same trend, but it is still clearly visible that the lake sediment has a much broader range in isotopic values than the deeper talik. In Fig. 6b we can show that the isotope data do not follow a clear trend anymore as in Fig. 5b and are therefore not primarily driven by SOM content (L530) which is an important distinction from Fig. 5b.

We had not plotted $\delta^{I3}C_{DOC}$ against DOC_{norm} yet but were curious to see how they would correlate (please see graph below). The lake mud (brown) and the deeper talik (blue) are mostly separated in two clusters. While the lake mud still appears to have a trend of increasing carbon isotope values with higher DOC amount the deeper talik appears to be mostly unaffected by changes in DOC_{norm} . Also, there is no clear trend anymore across the whole sediment core profile (R^2 =0.0115) as seen in Fig. 5b.



7.1505. DOC is not highly bioavailable, especially when it's allochtonous material. It's of course a relative measure, but have a look at the Catalan paper on half-lives of DOM along the aquatic continuum. In boreal systems, oligotrophic, cold, terrigenic DOM can be quite resistant to degradation.

In our manuscript we argue that the DOC present in the sediment porewater is mainly formed in-situ in the sediment itself and not brought in through hydrological pathways based on the results summarized in Fig. 5a. While the SOC in deeper talk layers and the bulk of the lake sediment are indeed of allochthonous (terrestrial) origin, the porewater DOC appears to be mostly formed within the sediment layers itself. We very much agree that not all DOC

compounds are highly bioavailable as we also pointed out in the discussion. We will modify the concerning sentence in L500-501 to make that point about the bioavailability of DOC more clearly:

"This assumption is also based on the fact that <u>certain_DOC compounds can be</u> is highly bioavailable..." The Catalan et al. (2016) paper also raises the important point of decreasing OC decomposition rates with increased water retention times due to the loss of its most reactive components in the early decomposition processes and an enrichment of the carbon pools with compounds that are degraded more slowly.

We also raised the same point in L508-510 of the manuscript: 'Preferential removal of the most labile carbon compounds such as simple polysaccharides which are naturally enriched in ^{13}C lead to an accumulation in less degradable compounds like lignin... '. In L510 we will add the Catalan et al. (2016) paper as an additional and important reference as well as in the list of references (L659).

8. Kudos for discussing pH dependence solubility and sorption/desorption of DOM. *Thank you for the kind acknowledgement.*