

Response to comments by reviewer 1 on cp-2024-60

We would like to thank Reviewer 1 for the meticulous review and valuable suggestions, which helped us to improve this manuscript significantly. Below, we address each of Reviewer 1's comments in detail. The reviewer's comments are in blue, and our responses are in black.

In this manuscript, Lin et al., evaluate changes in terrestrial organic matter input into the Arctic ocean over the last 18,000 years. The chosen study sites drain vast quantities of permafrost, such that enhanced terrestrial OC input might indicate enhanced permafrost thaw. The authors identify several pulses of terrestrial OC burial, some of which may correlate to known climatic events (e.g. meltwater pulse 1a). There is remarkably low terrOC input during the Holocene but lots of variability in terrOC delivery between 10 to 16 kyr. Each peak shows different compositional characteristics, suggesting distinct terrOM sources derived from different mechanisms. However, it would be interesting to try and unravel this further using your existing data (see comments below). Overall, the manuscript is well written, the figures are clear and the captions offer an appropriate level of detail. A few comments are included below that would be worth exploring further...

Comment 1: it is hard to tell whether the pulse in terrestrial OC is due to permafrost thaw or enhanced delivery of plant/soil OC. This could be assessed by measuring ^{14}C values in different lipids, but I realize is beyond the scope of this paper. However, there is existing literature that could be helpful - for example, Feng et al. (2013; PNAS) explored how the ^{14}C signature of different compound classes varied across the Pan-Arctic region and lignin phenols appear to mostly derive from recent carbon, whereas n-alkanes are derived from older carbon sources (e.g. permafrost). Although you don't have ^{14}C measurements, it could imply distinct sources for lignin vs n-alkanes in your samples.

...but another way to tackle this could be to look at other n-alkane indices such as the carbon preference index. This is frequently used to assess changes in OC maturity and may provide further insights into the type of terrOC that is being delivered into the marine realm, especially during the three pulses. If it was older OC, it may yield slightly lower CPI values. It may also tell you whether you are reworking old petrogenic OC into the marine realm too (which could be a CO_2 source if it was oxidised; see work by Sparkes 2016 the Cryosphere, but also work by Bob Hilton/Valier Galy etc)

We appreciate Reviewer 1's suggestions. The CPI values in cores PS51/154 and PS51/159 (4.5–6.5) fall within the range typical for surface and deep permafrost (Wild et al., 2022; Sánchez-García et al., 2014), and remain significantly higher than CPI values from petrogenic sources (~ 1) (Bray and Evans, 1961). Degradation indices, such as CPI and the Sd/SI ratio, show only small variations during the peaks of terrOM accumulation (Figure A1-1; Figure 2 in manuscript). Instead, CPI ratios suggest that fresher OM was generally deposited during the deglaciation between around 16 and 11 kyr BP, likely due to shorter transport distances to the core site at lower sea levels. Lignin phenol $\Sigma 8$ / HMW fatty acid ratios are consistent with CPI trends, but the impact of the slumping layer is more pronounced (Figure A1-1). We will include CPI data in the supplementary plots and briefly discuss how transport distance to the core site is reflected in the biomarker data.

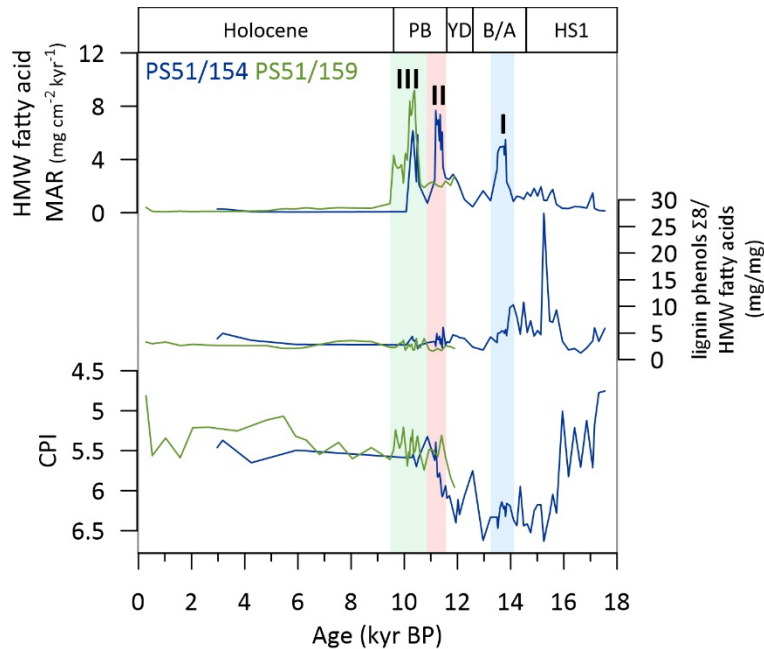


Figure A1-1. Mass accumulation rates (MARs) of high molecular weight (HMW) fatty acids, ratio of lignin phenol $\Sigma 8$ to HMW fatty acids contents, and the carbon preference index (CPI) in cores PS51/154 and PS51/159. Colored bars highlight periods of HMW fatty acid MAR peaks: from 14.1 to 13.2 kyr BP (blue, MAR peak I), from 11.6 to 10.9 kyr BP (red, MAR peak II), and from 10.9 to 9.5 kyr BP (green, MAR peak III).

Comment 2: Export vs preservation

It is important to confirm that the increase in leaf wax mass accumulation rates is not due to enhanced preservation but is reflecting enhanced terrOC export. This could be explored by calculating MARs of short-chain alkanes (algal-derived), mid-chain alkanes (moss or macrophyte derived), and long-chain alkanes (vascular plant) *n*-alkanes during the three “pulses”. If all three increase, it might suggest that OC preservation is the main driver. But if only the mid- and long-chain alkane MARs increase, it would imply enhanced terrOC input.

We thank Reviewer 1’s insight. However, disentangling the cause of elevated terrOM MAR peaks, whether due to enhanced terrOM export or enhanced preservation, is challenging. This difficulty arises mainly because marine primary production likely increased during periods of elevated terrOM export. Terrestrial nutrients serve as an important source to fuel marine primary production in the Arctic Ocean (Terhaar et al., 2021), and elevated terrOM export could stimulate marine primary production, leading to simultaneous increases in terrestrial and marine OM. Secondly, terrOM MAR peaks in cores PS51/154 and PS51/159 coincided with periods of low seasonal sea ice cover (Hörner et al., 2016) (Figure 2 in the manuscript). Reduced sea ice cover favors phytoplankton blooms, while extended open-water conditions provide more area and time for phytoplankton growth (Shiozaki et al., 2022), further increasing marine primary production. The contents per gram of OC (Figure A1-2) and MARs (Figure A1-3) of short-chain, mid-chain, and long-chain *n*-alkanes show no significant differences among the three homologues.

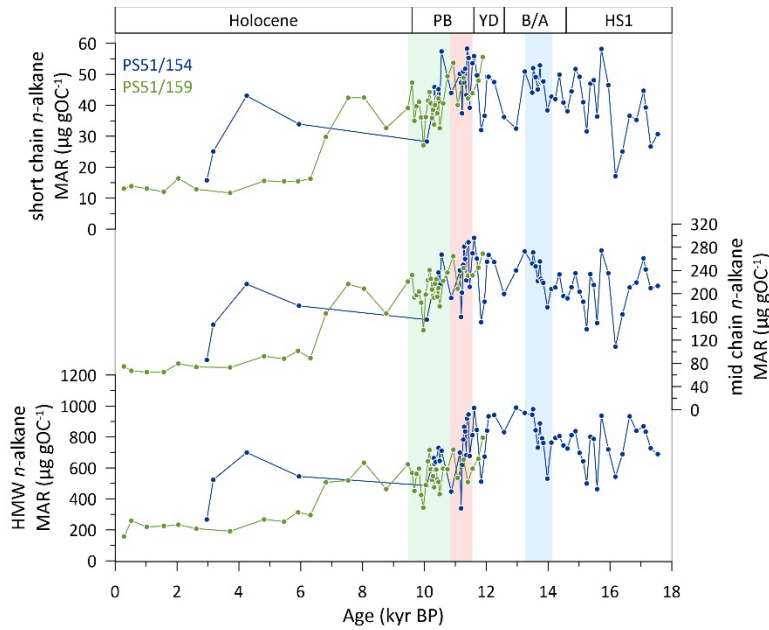


Figure A1-2. Contents of short-chain, mid-chain, and long-chain (HMW) *n*-alkanes in cores PS51/154 (dark blue) and PS51/159 (light green). Colored bars highlight periods of HMW fatty acid MAR peaks: from 14.1 to 13.2 kyr BP (blue), from 11.6 to 10.9 kyr BP (red), and from 10.9 to 9.5 kyr BP (green).

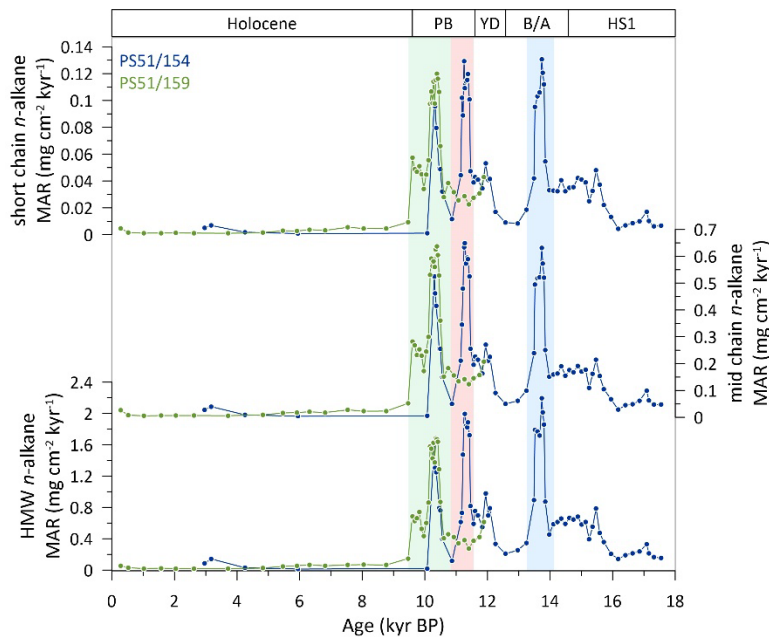


Figure A1-3. Mass accumulation rates (MARs) of short-chain, mid-chain, and long-chain (HMW) *n*-alkanes in cores PS51/154 (dark blue) and PS51/159 (light green). Colored bars highlight periods of HMW fatty acid MAR peaks: from 14.1 to 13.2 kyr BP (blue), from 11.6 to 10.9 kyr BP (red), and from 10.9 to 9.5 kyr BP (green).

Comment 3: a note on pAq

The authors use the pAq index (the ratio between mid vs long chain *n*-alkanes) to infer changes in wetland input, but I would note that's its more complex than this. For example, both Sphagnum moss and aquatic

macrophytes are characterized by similar lipid distributions (Baas et al., 2000; Ficken et al., 2000), so without knowledge of the local vegetation, it's challenging to say whether the P_{aq} ratio is due to changes in moss input or macrophyte input. Perhaps you could narrow this down by drawing upon predicted vegetation patterns during the Holocene/LGM etc. There is also great work by Jorien Vonk, Bart van Dongen, Orjan Gustafsson etc in similar pan-arctic regions that might be useful. For example, Vonk 2009 paper in *Org. Geochem* shows that "...the $C_{25}/(C_{25} + C_{29})$ *n*-alkane ratio is most suitable for terrestrial OM source apportionment in these coastal regions". This might be worth exploring alongside the P_{aq} (although I suspect you will get similar results!).

We thank Reviewer 1's suggestion and for kindly pointing us to the relevant references. For the first part of the suggestion, we will include references on predicted moss cover in Siberia during the last deglaciation (Andreev et al., 2003; Hubberten et al., 2004) and in recent times (Van Dongen et al., 2008). For the second part of the suggestion, we compared the $C_{25}/(C_{25}+C_{29})$ *n*-alkane ratio with P_{aq} (Figure A1-4). While both indices yield similar results, the $C_{25}/(C_{25}+C_{29})$ *n*-alkane ratio better reflects moss/peatland input and is less influenced by species and temperature variations (Vonk and Gustafsson, 2009; Van Dongen et al., 2008). Accordingly, we will adopt the $C_{25}/(C_{25}+C_{29})$ *n*-alkane ratio in the revised manuscript, retaining the same interpretations.

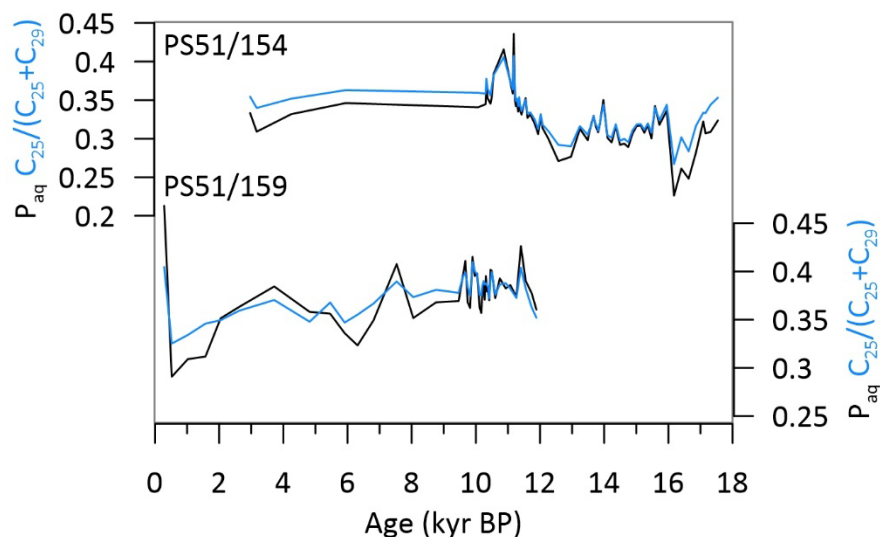


Figure A1-4. P_{aq} (black lines) and $C_{25}/(C_{25}+C_{29})$ (blue lines) indices for cores PS51/154 and PS51/159 (this study).

Minor comments:

L63: v/v – the v 's should be italicised – change throughout

We will correct this in the revised manuscript.

L205: correct that it's used for macrophytes – but also for sphagnum mosses.

We appreciate reviewer 1 for pointing this out, in the revised manuscript we will replace P_{aq} index with the $C_{25}/(C_{25}+C_{29})$ index and replace this sentence with an introduction of the $C_{25}/(C_{25}+C_{29})$ index.

L252: is this statistically significant?

During 16.2 and 9.5 kyr BP, both $\delta^{13}\text{C}$ and TOC in cores PS51/154 and PS51/159 show no correlation with HMW fatty acid MAR, as evidenced by low R and high p -values (Figure A1-5). To improve clarity, we will revise the sentence of L252 as follows: “The values of $\delta^{13}\text{C}$ and TOC in cores PS51/154 (–25.6‰ and 0.79%) and PS51/159 (–25.8‰ and 0.99%) remained rather constant between 16.2 and 9.5 kyr BP, despite the three periods of peak MAR of terrestrial biomarkers (Fig S5).”.

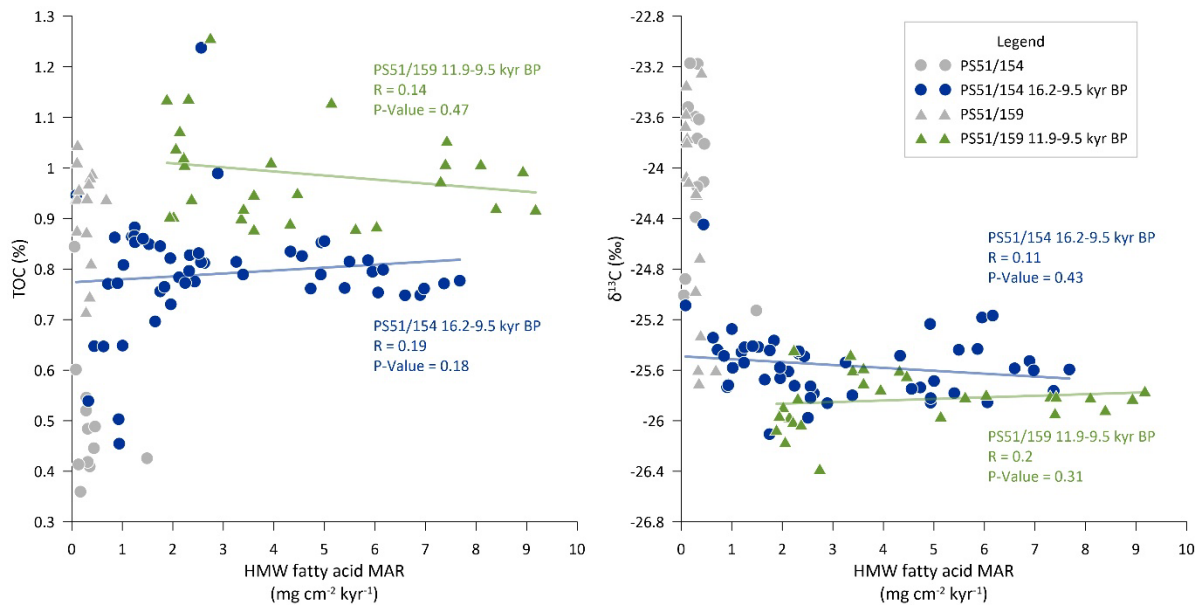


Figure A1-5. TOC and $\delta^{13}\text{C}$ versus high molecular weight (HMW) fatty acid mass accumulation rate (MAR) in cores PS51/154 and PS51/159. Data points from 16.2 to 9.5 kyr BP are highlighted and the linear regression lines are shown. The low correlation and high p -value indicate no significant changes in TOC and $\delta^{13}\text{C}$ values did not change during this period despite elevated terrestrial organic matter inputs.

L365: dialkyl

Please refer to the next reply.

L366: and also found in peats – and lakes - and marine sediments!

We appreciate the reviewer’s meticulous review. The original sentence from L365 and 366 is “Branched Glycerol Dialkyl Glycerol Tetraethers (brGDGTs) are found in membrane lipids from soil bacteria.” We agree that brGDGTs are also found in aquatic and marine environments. This original sentence in L365-366 will be removed, as the brGDGT MAR record of cores SO202-18-3/6 (Meyer et al., 2019) is not from the Arctic Ocean and may not reflect comparable environmental conditions.

L412: subscript CO₂

We will correct the typo in the revised manuscript.

References

- Andreev, A. A., Tarasov, P. E., Siegert, C., Ebel, T., Klimanov, V. A., Melles, M., Bobrov, A. A., Dereviagin, A. Y., Lubinski, D. J., and Hubberten, H. W.: Late Pleistocene and Holocene vegetation and climate on the northern Taymyr Peninsula, Arctic Russia, *Boreas*, 32, 484-505, 2003.
- Bray, E. and Evans, E.: Distribution of n-paraffins as a clue to recognition of source beds, *Geochimica et Cosmochimica Acta*, 22, 2-15, 1961.
- Hörner, T., Stein, R., Fahl, K., and Birgel, D.: Post-glacial variability of sea ice cover, river run-off and biological production in the western Laptev Sea (Arctic Ocean)—A high-resolution biomarker study, *Quaternary Science Reviews*, 143, 133-149, 2016.
- Hubberten, H. W., Andreev, A., Astakhov, V. I., Demidov, I., Dowdeswell, J. A., Henriksen, M., Hjort, C., Houmark-Nielsen, M., Jakobsson, M., Kuzmina, S., Larsen, E., Lunkka, J. P., Lysa, A., Mangerud, J., Möller, P., Saarnisto, M., Schirmer, L., Sher, A. V., Siegert, C., Siegert, M. J., and Svendsen, J. I.: The periglacial climate and environment in northern Eurasia during the Last Glaciation, *Quaternary Science Reviews*, 23, 1333-1357, 10.1016/j.quascirev.2003.12.012, 2004.
- Meyer, V. D., Hefter, J., Köhler, P., Tiedemann, R., Gersonde, R., Wacker, L., and Mollenhauer, G.: Permafrost-carbon mobilization in Beringia caused by deglacial meltwater runoff, sea-level rise and warming, *Environmental Research Letters*, 14, 10.1088/1748-9326/ab2653, 2019.
- Sánchez-García, L., Vonk, J. E., Charkin, A. N., Kosmach, D., Dudarev, O. V., Semiletov, I. P., and Gustafsson, Ö.: Characterisation of Three Regimes of Collapsing Arctic Ice Complex Deposits on the SE Laptev Sea Coast using Biomarkers and Dual Carbon Isotopes, *Permafrost and Periglacial Processes*, 25, 172-183, 10.1002/ppp.1815, 2014.
- Shiozaki, T., Fujiwara, A., Sugie, K., Nishino, S., Makabe, A., and Harada, N.: Bottom-associated phytoplankton bloom and its expansion in the Arctic Ocean, *Global Change Biology*, 28, 7286-7295, 10.1111/gcb.16421, 2022.
- Terhaar, J., Lauerwald, R., Regnier, P., Gruber, N., and Bopp, L.: Around one third of current Arctic Ocean primary production sustained by rivers and coastal erosion, *Nature Communications*, 12, 169, 10.1038/s41467-020-20470-z, 2021.
- van Dongen, B. E., Semiletov, I., Weijers, J. W. H., and Gustafsson, Ö.: Contrasting lipid biomarker composition of terrestrial organic matter exported from across the Eurasian Arctic by the five great Russian Arctic rivers, *Global Biogeochemical Cycles*, 22, 10.1029/2007gb002974, 2008.
- Vonk, J. E. and Gustafsson, Ö.: Calibrating n-alkane Sphagnum proxies in sub-Arctic Scandinavia, *Organic Geochemistry*, 40, 1085-1090, 10.1016/j.orggeochem.2009.07.002, 2009.
- Wild, B., Shakhova, N., Dudarev, O., Ruban, A., Kosmach, D., Tumskoy, V., Tesi, T., Grimm, H., Nybom, I., Matsubara, F., Alexanderson, H., Jakobsson, M., Mazurov, A., Semiletov, I., and Gustafsson, Ö.: Organic matter composition and greenhouse gas production of thawing subsea permafrost in the Laptev Sea, *Nature Communications*, 13, 5057, 10.1038/s41467-022-32696-0, 2022.