Anonymous Referee #2

The authors present a multi-proxy study using $\delta^{13}C$, $\delta^{15}N$, C/N ratios, and terrestrial n-alkane biomarkers from surface sediments and sediment cores to reconstruct terrestrial organic carbon (OC) inputs to the Chukchi Sea over the last three centuries compared with published datasets on temperature, sea ice and precipitation. This work contributes to a growing body of literature documenting the land-ocean carbon fluxes in the Arctic and the role of sea ice and climate forcing in shaping sedimentary OC signatures. However, several significant methodological and interpretive issues must be addressed to strengthen the manuscript's validity and clarity. As written, the manuscript needs substantial work to be publication-quality and contribute to advancing our understanding of these topics.

We would like to thank anonymous Referee #2 for the comments and suggestions.

A few major issues that need to be addressed:

Title and Abstract

The abstract summarizes results but overstates interpretive certainty. Please consider softening the statement "provide evidence of a significant link..." to reflect that this is an inferred relationship based on proxy correlation, not direct causality.

In line 290 we changed the sentence to "provide a link between sea ice and terrestrial OC export…"

Line 18 – "the response of the Arctic", please be more specific here. Response of marine primary production or biological pump in Arctic marine ecosystems

This sentence has been modified to "yet environmental changes in the Arctic (temperature, precipitation, and sea ice) are not well characterized and fully understood".

Line 24 – from increased fluvial sources

Yes. This is confirmed by the significant correlation between TERR-alkanes data from the LV77-4 core and mean annual discharge from Lena River reflecting the influence of fluvial sources on the terrestrial organic carbon content of the sediments in the region (see Table 1 below showing correlation of biomarkers against climatic parameters).

Line 25 – Not entirely convinced that air temperature and sea ice are the key role here but rather increased precipitation, runoff from boreal rivers, and seasonal pulses from the coastal currents.

Our new comparison between biomarker indexes from our five cores and climatic parameters reveals that only data from LV77-4 indicate significant correlation with Lena River discharge (see Table 1 & Figure 4), the major conduit of land-derived material to the sea and then across the shelf via the Siberian Coastal Current (SCC). Although biomarker indexes also show significant correlation with annual precipitation from Chukotka, this later is relatively low (250-300 mm). The river discharge also involves snow and glaciers melt as a result of the rising surface air temperature and related melting of the permafrost. On the other hand, biomarker data from core 14S03, the other coastal core northern of Alaska, only show significant correlation with air temperature and sea ice cover while influence from the more distant Yukon River seems to be negligible.

Line 35 – The most recent Arctic Report Card (2024) indicates a shift of the Arctic permafrost as a source rather than a sink. Relevant to include/discuss.

Agreed

Line 40 - There is a more recent and relevant article indicating a third to half of NPP in the AO is from rivers and coastal erosion. This work should be considered and incorporated here:

Terhaar, J., Lauerwald, R., Regnier, P. et al. Around one third of current Arctic Ocean primary production sustained by rivers and coastal erosion. Nature Comm, 12, 169 (2021). https://doi.org/10.1038/s41467-020-20470-z

This study has been added in the revised version of the manuscript.

Line 41 – last few decades? Please be more specific.

Last two decades

Line 43 – Arrigo has done substantial work on increasing NPP in the Arctic. The authors may want to consider including prior work (e.g. Arrigo and van Djiken 2015 https://doi.org/10.1016/j.pocean.2015.05.002)

Agreed

Methods

The authors state surface sediments were collected, but do not indicate how they were collected (e.g., Van Veen grab, box core, top of multi-corer). This is critical for understanding whether the samples represent intact, undisturbed surface material suitable for modern flux interpretation.

The uppermost 0-2 cm sediments were collected with a box-corer. This is now in the revised version.

It is unclear whether an internal laboratory standard was used in addition to the IAEA-certified reference materials. Internal standards are essential for monitoring instrument drift and analytical reproducibility. Please clarify.

There seems to be confusion between biomarker and bulk isotope determinations. For biomarker we didn't use an internal but external standard for quantification which is the 5α-cholestane that was added prior GC-MS analysis.

For $\delta^{13}C$ and $\delta^{15}N$ of bulk organic matter we used IAEA-certified reference materials as described in the method section of the paper (2.2 Bulk analyses). There is no addition of any internal standard for bulk measurements.

The methods mention the addition of 5α-cholestane but make no reference to derivatization (e.g., silylation using BSTFA). Derivatization is typically required for lipid compound analysis via GC-MS, particularly if alcohols or acids were present. Please clarify whether derivatization was performed and, if not, justify its omission.

5α-cholestane is a cyclic hydrocarbon, with no OH function, which thus does not need to be derivatized.

Results and Discussion

While the authors report measurements of total nitrogen (TN) and $\delta^{15}N$ in sediments, these data are not meaningfully discussed in the manuscript. It remains unclear how these variables contribute to the study's central questions or interpretations. $\delta^{15}N$ is a valuable tracer of nitrogen sources and cycling processes (e.g., denitrification, organic matter origin), and TN content can inform on organic matter input and preservation. I strongly encourage the authors to either integrate these measurements into their broader discussion—particularly in relation to sediment diagenesis, productivity, or regional biogeochemical shifts—or clarify why they were measured if they are not central to the narrative. As it stands, the inclusion of these variables without interpretation detracts from the overall coherence of the manuscript.

We added some discussion on TN and $\delta^{15}N$ to account for denitrification which is found to occur in the Chukchi Sea mainly in sediments (as water column is well oxygenated) and to induce huge loss of nitrate making this region highly N-limited during the phytoplankton growth season (Hardison et al., 2017). Chang and Devol (2009) have found based on whole-core incubations that denitrification is higher in the shallow than deep water-sediments of the Chukchi Sea. Recently, Brown et al. (2015) have shown that nitrification (transformation of ammonium to nitrate) that occurs in the bottom waters of the Chukchi Sea is responsible for significant ^{15}N enrichment of nitrate ($\delta^{15}N$ varies between 3.5 and 9.5). Our data indicate high $\delta^{15}N$ values in all records ranging from 5.5 to 9.5 which probably reflect partial nitrification-denitrification process as described in Brown et al. (2015).

Degradation of permafrost represents another factor responsible for the release of huge amount of dissolved nitrogen and nitrate with heavy $\delta^{15}N$ signature of 7 and 12 ‰, respectively, mostly due to the denitrification of nitrate released from the degraded permafrost (Francis et al., 2023). However, according to these authors, this is only found in local site of Kolyma River where permafrost degradation is high and the recorded heavy signal of $\delta^{15}N$ from this site is found to be lost in the rest of the riverine and estuarine zones (with $\delta^{15}N$ values ranging between 2 and 5 ‰). Therefore, this factor is unlikely to account for the heavy $\delta^{15}N$ records from our sediment cores.

Hardison, A. K., McTigue, N. D., Gardner, W. S., & Dunton, K. H. (2017). Arctic shelves as platforms for biogeochemical activity: Nitrogen and carbon transformations in the Chukchi Sea, Alaska. Deep Sea Research Part II: Topical Studies in Oceanography, 144, 78-91.

Chang, B. X., & Devol, A. H. (2009). Seasonal and spatial patterns of sedimentary denitrification rates in the Chukchi Sea. Deep Sea Research Part II: Topical Studies in Oceanography, 56(17), 1339-1350.

Brown, Z. W., Casciotti, K. L., Pickart, R. S., Swift, J. H., & Arrigo, K. R. (2015). Aspects of the marine nitrogen cycle of the Chukchi Sea shelf and Canada Basin. Deep Sea Research Part II: Topical Studies in Oceanography, 118, 73-87.

Francis, A., Ganeshram, R. S., Tuerena, R. E., Spencer, R. G., Holmes, R. M., Rogers, J. A., & Mahaffey, C. (2023). Permafrost degradation and nitrogen cycling in Arctic rivers: insights from stable nitrogen isotope studies. Biogeosciences, 20(2), 365-382.

The proposed linkage between $ACL_{(27–31)}$ and evapotranspiration/drier conditions is speculative and not well-supported. ACL typically reflects changes in source vegetation or degradation, not moisture balance directly. Please revise this interpretation or support it with more appropriate references (e.g., compound-specific isotope data or vegetation studies).

Odd high molecular weight n-alkanes are constituents of epicuticular waxes and ACL_{27-31} have been for quite long period used by the biomarker community as a proxy of moisture availability which in this region is tightly related to temperature and vegetation type. There has been many publications that used this approach, which we included in the revised version of the paper (Bush and McInerney 2013; 2015; He et al., 2019; Andersson et al., 2011).

Bush, R. T., & McInerney, F. A. (2015). Influence of temperature and C4 abundance on n-alkane chain length distributions across the central USA. Organic Geochemistry, 79, 65-73.

He, D., Huang, H., & Arismendi, G. G. (2019). n-Alkane distribution in ombrotrophic peatlands from the northeastern Alberta, Canada, and its paleoclimatic implications. Palaeogeography, Palaeoclimatology, Palaeoecology, 528, 247-257.

Andersson, R. A., Kuhry, P., Meyers, P., Zebühr, Y., Crill, P., & Mörth, M. (2011). Impacts of paleohydrological changes on n-alkane biomarker compositions of a Holocene peat sequence in the eastern European Russian Arctic. Organic Geochemistry, 42(9), 1065-1075.

Bush, R. T., & McInerney, F. A. (2013). Leaf wax n-alkane distributions in and across modern plants: Implications for paleoecology and chemotaxonomy. Geochimica et Cosmochimica Acta, 117, 161-179.

Although precipitation trends are mentioned and included in the supplementary materials, the authors dismiss the lack of correlation without much exploration. Given that precipitation and runoff are primary vectors for terrestrial OM export, more effort should be made to assess their potential role. Data from R-ArcticNET could be one option to explore. Additionally, the role of river discharge is sorely missing. The inclusion of Walsh's sea ice reconstructions was interesting but there is something not quite connecting

In the revised version of our manuscript, we evaluated the correlation between TERR-alkanes and related indexes from each core against river discharges from the main rivers of Alaska (Yukon & Mackenzie) and Siberia (Lena & Kolyma) as well as instrumental/paleo temperature, sea ice concentration and precipitation data. All correlation calculations are provided in Table 1. Correlations considered significant at $p \leq 0.05$ are further discussed in the manuscript. Our results highlight the key role of Lena River discharge together with temperature and sea ice cover to account for the terrestrial content of LV77-4 while in the eastern Chukchi Sea at site 14S03, only temperature and sea ice cover are driving factors, presumably reflecting minor role of rivers as compared to the core close to the East Siberian Sea. Figures 4 & 5 have been modified accordingly.

While the discussion focuses on a potential link between the Siberian Coastal Current and the delivery of terrestrial biomarkers (e.g., long-chain n-alkanes and % sphagnum moss), it fails to adequately address the lack of observed relationships with the Alaska Coastal Current (ACC). This is a critical omission, especially given the ACC's strong seasonal influence on shelf circulation and potential for transporting terrestrial material from Alaskan riverine sources. A more comprehensive analysis comparing the spatial trends of biomarker distributions with known current trajectories (e.g., ACC vs. SCC) would strengthen the interpretation and may reveal key regional differences in sediment provenance and deposition. Existing circulation models and observational studies (e.g., Danielson et al. 2017; Woodgate 2018) could provide useful context. Other studies focusing on riverine inputs (e.g., changes in Yukon River discharge) could also be considered more substantively (e.g., Bennett et al. 2015 https://doi.org/10.1016/j.jhydrol.2015.04.065)

As mentioned in the response to the previous comment, we distinguish between a dominant influence of riverine discharge on the terrestrial content of LV77-4, presumably through the SCC, and prevalence of temperature and sea ice at 14S03 core. We further discuss these features in the revised version of our manuscript.

The discussion does not address bioturbation at all, which is known to affect Pb-210 and Cs-137 profiles in this shallow, highly productive, continental shelf region. Bioturbation can substantially obscure the vertical profile of unsupported 210Pb in Arctic shelf and slope environments. Given that the study region is known to experience intense bioturbation by benthic fauna — as documented in prior work (e.g., Cooper and Grebmeier, 2018, https://doi.org/10.1016/j.dsr2.2018.01.009). The assumption of a steady-state depositional model may be inappropriate or at least requires additional justification. I recommend the authors discuss whether signs of mixing (e.g., non-exponential decay profiles, homogenized surface layers) were observed and, if possible, support their dating interpretation with corroborating tracers such as 137Cs (which is briefly mentioned but not reported, line 72).

All age models used here have been previously published (Bai et al., 2022; Su et al., 2023; Jalali et al., 2024). A discussion on the age models is provided in the aforementioned studies especially regarding the post-depositional processes such as bioturbation and physical reworking of sediments. Based on ^{137}Cs and ^{210}Pb profiles there was no evidence of bioturbation or physical reworking, and therefore these cores were selected for further chemical analyses.

Bai Y, Sicre M A, Ren J, Jalali B, KleinV, Li H and Chen J: Centennial-scale variability of sea-ice cover in theChukchi Sea since AD 1850 based on biomarker reconstruction Environ. Res. Lett. 17 044058, 2022.

Jalali, B., Sicre, M. A., Ren, J., Klein, V., Li, Z., Su, L., ... & Chen, J.: Reconstruction of sea ice variability in the Chukchi Sea during the last three centuries based on biomarker proxies. Environmental Research Communications, 6(9), 091013, 2024.

Su L., Ren J., Sicre M. A., Bai Y., Zhao R., Han X. and Chen J.: Changing sources and burial of organic carbon in the Chukchi Sea sediments with retreating sea ice overrecent centuries. Climate of the Past 19 1305–20, 2023.

Conclusions

The statement that the record shows "strong terrestrial OC export events in pre-industrial times" needs to be tempered unless the dating uncertainties and bioturbation issues are directly addressed. Also, reinforce that ACL interpretations are tentative given the current uncertainty about their linkage to hydrologic regime. Consider expanding the conclusion to explicitly reflect the uncertainties in proxy interpretations (ACL, Paq, % Sphagnum) and their implications for reconstructing Arctic terrestrial OC dynamics.

As we mentioned above, dating uncertainties, bioturbation and physical reworking at our core sites are negligible. We also provided some relevant references reinforcing the usefulness of ACL_{27-31} as a proxy for past hydroclimate/ temperature changes.

In general, the paper also lacks some relevant references. I've included a few but suggest the authors do another thorough review of work that has been done in the region.

We added references to the revised version.

Other Minor/Specific Comments

Line 93 – The authors used 2-4 g of sediment. This seems like a lot. Is there a published method the authors were following? Perhaps in the Bai or other Jalali paper?

It's not unusual to extract 2 to 4 g of sediments for biomarkers in particular to ensure the detection of highly branched isoprenoid alkenes (IP25, HBI-II and HBI-III).

Line 127 – TEER should be TERR

Done

Line 143-44 – This statement is unclear and inaccurately described. All records are decreasing ? or are they more/less enriched or depleted. The latter language should be used to describe changing isotopic values. Additionally, it is unclear why there is a focus on 1850-1900. Why this short 50 year window? Do the data support this resolution of analysis? As stated above, there are some concerns about bioturbation that need to be addressed.

This sentence has been rephrased to "All cores depict a depleting trend since approximately 1850-1900 towards the end of the records except for the northernmost station R1 where they show continuous enrichment of more depleted values".

1850-1900 is not the time-period during which the described changes are seen but rather the time from which the $\delta^{13}C$ started to decline/increase towards the end of the five records (starting change is not simultaneous in the five records).

Line 242 – Figure 4: The scale for sea ice concentration is not appropriate. Please limit the scale to 0–100%.

Done in the revised version of Figure 4 (figure enclosed).

Line 290 – statistically significant, or just "significant"?

It's statistically significant based on our correlations (Table 1).

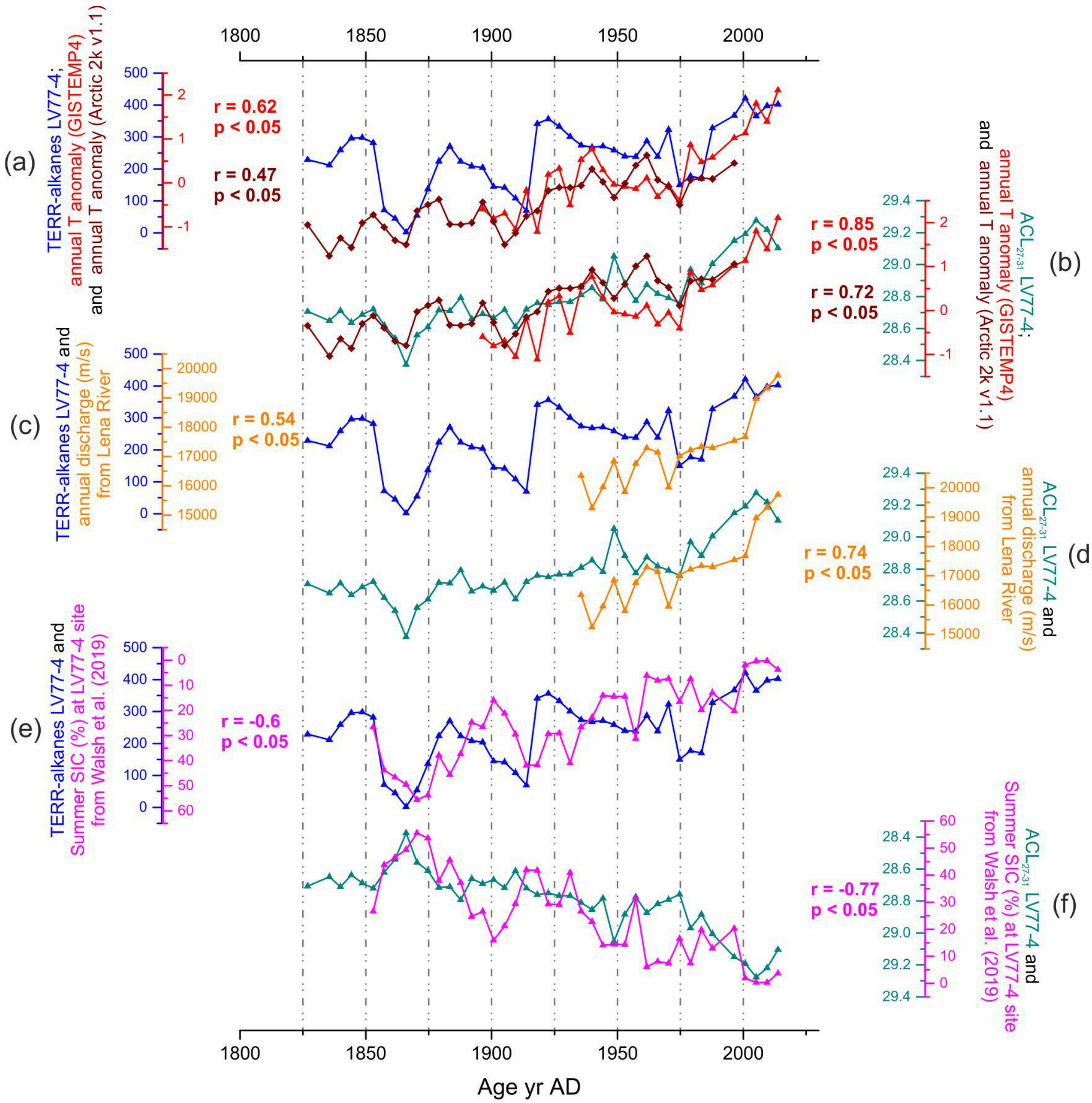

Figure 4: (a) Comparison of the TERR-alkanes C27-C31 concentration (in µg/g TOC) obtained from core LV77-4 with average air temperature anomalies (°C) over Alaska and Chukotka (shown in red; https://data.giss.nasa.gov/gistemp/) and Arctic air temperature anomaly (°C) from PAGES 2k consortium (2013; shown in dark red). (b) Same as (a) but for ACL_{27-31}. (c) Comparison of the TERR-alkanes C27-C31 concentration (in µg/g TOC) obtained from core LV77-4 with mean annual discharge (m/s) from Lena River (https://arcticgreatrivers.org/discharge/). (d) Same as (c) but for ACL_{27-31}. (e) Comparison of the TERR-alkanes C27-C31 concentration (in µg/g TOC) obtained from core LV77-4 with historical and satellite summer sea ice

concentration (%) at the same core site from Walsh et al. (2019). (f) Same as (e) but for ACL_{27-31}. Pearson's r for each comparison is also provided.

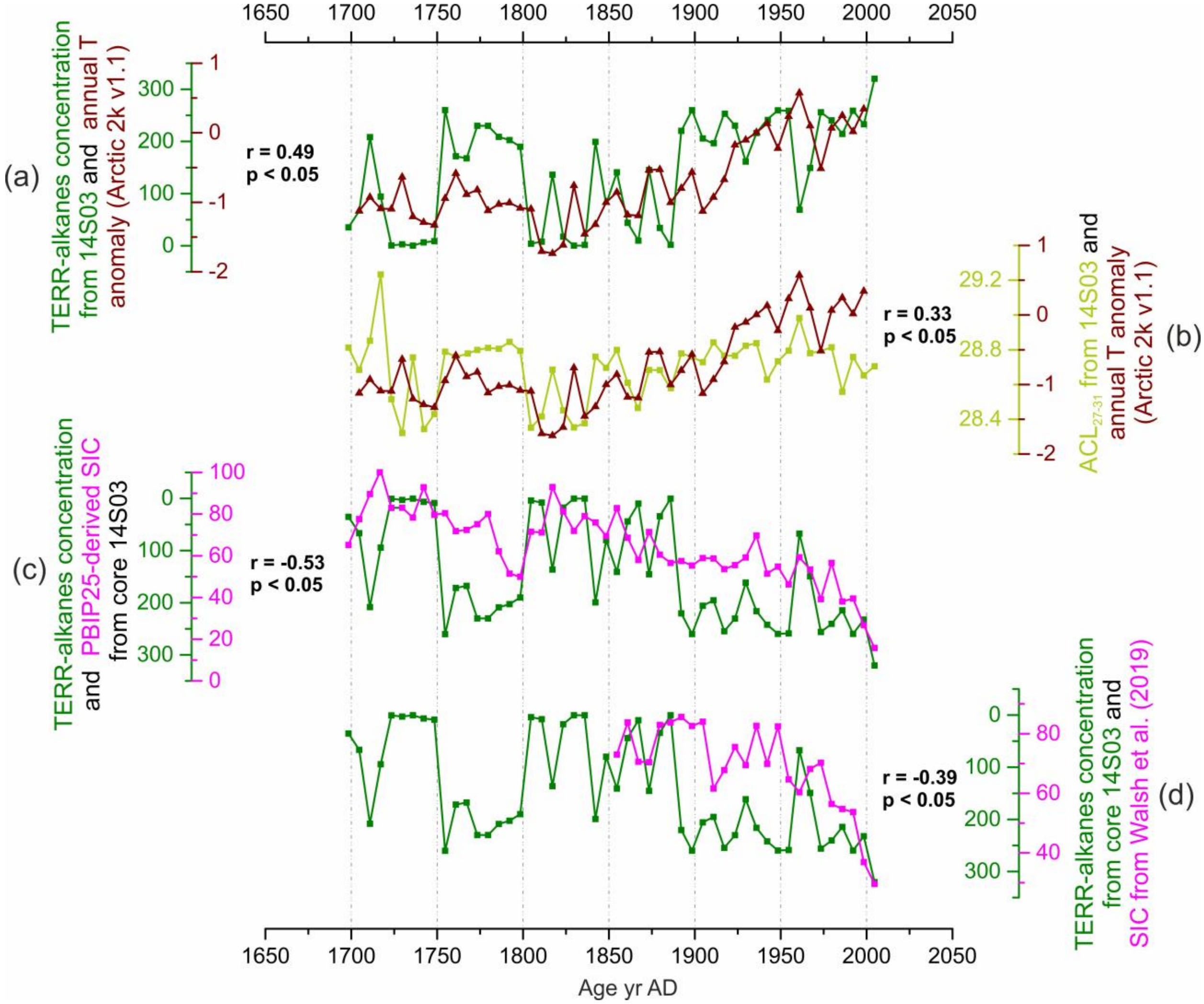

Figure 5: (a) Comparison of the TERR-alkanes C27-C31 concentration (in µg/g TOC) obtained from core 14S03 (in green) with Arctic annual air temperature anomaly (°C) from PAGES 2k consortium (2013; shown in red). (b) Same as (a) but for ACL_{27-31}. (c) Comparison of the TERR-alkanes C27-C31 concentration (in µg/g TOC; inversed Y axis) obtained from core 14S03 (in green) with P_BIP_{25}-derived sea ice concentration reconstructed from the same core using the calibration of Jalali et al. (2024). (d) Same as (c) but for comparison with historical and satellite sea ice concentration from Walsh et al. (2019) at 14S03 core site. Pearson's r for each comparison is also provided.

Tables 1: Table showing correlation coefficient (Pearson's r) calculated between TERR-alkanes, ACL_{27-31} and P_{aq} from each core and climatic parameters (temperature, precipitation, river discharge and sea ice concentration). Correlation is considered significant at $p \leq 0.05$ (shown in bold). Instrumental surface air temperature are from GISTEMP4 (https://data.giss.nasa.gov/gistemp/) while Arctic paleo-temperature are from PAGES 2k consortium (2013). Average annual precipitation data from Alaska and Chukotka are from Global Precipitation Climatology Centre (GPCC; https://psl.noaa.gov/data/gridded/data.gpcc.html). Data of annual discharge from the four rivers (Lena, Kolyma, Yukon and Mackenzie) are from ArcticGR (https://arcticgreatrivers.org/discharge/). Historical and satellite data of summer (JAS) sea ice concentration (SIC) at each core site are from Walsh et al. (2019) while paleo-records of SIC from each core are calculated from $PBIP_{25}$ index using the calibration of Jalali et al. (2024).

	LV77-4			14S03			C007			R01			14R09		
	TERR-alkanes	ACL_{27-31}	P_{aq}	TERR-alkanes	ACL_{27-31}	P_{aq}	TERR-alkanes	ACL_{27-31}	P_{aq}	TERR-alkanes	ACL_{27-31}	P_{aq}	TERR-alkanes	ACL_{27-31}	P_{aq}
Instrumental T averaged over Alaska & Chukotka	**0.62**	**0.85**	**-0.86**	0.3	-0.34	0.4	0.09	0.09	-	-0.22	-0.09	0.15	-	-	-
Paleo T from the pan Arctic region	**0.47**	**0.72**	**-0.76**	**0.49**	**0.33**	-0.25	0.17	**0.57**	-	0.49	0.4	**-0.69**	-0.35	0.28	-0.39
Annual precipitation averaged over Alaska & Chukotka	**0.57**	**0.38**	**-0.45**	0.03	0.01	0.04	-0.44	-0.18	-	0.09	0.35	-0.29	-	-	-
Annual discharge of Lena River	**0.54**	**0.74**	**-0.83**	0.01	0.23	-0.19	**-0.81**	-0.33	-	-0.68	-0.23	0.5	-	-	-
Annual discharge averaged from Lena, Kolyma, Yukon and Mackenzie rivers	0.23	-0.01	0	-0.22	0.32	-0.4	0.11	0.37	-	-0.29	-0.08	0.25	-	-	-
Historical and satellite SIC (%)	**-0.6**	**-0.77**	**0.79**	**-0.39**	-0.02	0.05	0.2	0.49	-	-0.08	0.09	0.29	-0.13	-0.75	0.76
P_BIP_{25}-derived SIC (%)	-0.21	**-0.5**	**0.47**	**-0.53**	-0.11	0.11	0.23	-0.18	-	-0.08	0.12	0.09	**-0.82**	-0.34	0.18