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Key Points:

- Permafrost turnover and export can be traced using alkanolic acid $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ whereas bulk organic carbon (OC) isotope values are biased by coal-derived OC
- Alkanolic acid turnover in permafrost is multi-millennial likely controlled by low mean annual air temperature and precipitation
- Long-chain alkanolic acid $\Delta^{14}\text{C}$ values in river and fjord sediments imply reburial of deep active layer and permafrost OC

Supporting Information:

- Supporting Information S1
- Table S1
- Table S2

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Permafrost Organic Carbon Turnover and Export Into a High-Arctic Fjord: A Case Study From Svalbard Using Compound-specific ^{14}C AnalysisStephanie Kusch^{1,2,5} , Janet Rethemeyer³, Daniela Ransby^{1,4} , and Gesine Mollenhauer^{1,2}

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Abstract In a warming climate, thawing permafrost soils in the circumpolar Arctic region are subject to enhanced microbial turnover as well as mass mobilization and other erosion processes. High-Arctic settings such as Svalbard are exceptionally vulnerable to these effects, but the presence of coal deposits obscures the organic carbon (OC) signature of permafrost OC, particularly its carbon isotope composition, when studying OC turnover and export. Here, we analyze the compound-specific $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ isotopic composition of alkanolic acids from permafrost soils and river and fjord sediments to assess soil turnover in the catchment of the Bayelva River near Ny-Ålesund and trace transport and re-burial of permafrost OC into the adjacent Kongsfjord. Our data confirm the influence of coal-derived OC on $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of bulk soil and sedimentary OC, while alkanolic acid $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values are less affected by coal contributions. Alkanolic acid $\Delta^{14}\text{C}$ values in the soil profile imply long-term residence in soils prior to deposition in river and fjord sediments, that is, multi-millennial turnover that is significantly slower than reported from other environments. Strongly ^{14}C -depleted vascular plant-derived long-chain alkanolic acids can be found in Bayelva River and Kongsfjord sediments revealing substantial input of deep active layer/permafrost OC, particularly in the Bayelva River and off its river mouth. In the central Kongsfjord, long-chain alkanolic acid $\Delta^{14}\text{C}$ values are higher either reflecting input from other permafrost areas or physical effects resulting, for example, from deposition in settings with different accumulation rates or from sediment sorting.

Plain Language Summary Rising atmospheric temperatures have a particularly strong effect on carbon cycling in high latitude ecosystems such as Svalbard. Thawing of permanently frozen ground (permafrost) results in stronger microbial activity as well as erosion and reburial of previously frozen old carbon-rich material in aquatic systems. Such processes are poorly constrained in Svalbard and can be studied using carbon isotope analyses and ^{14}C dating. However, permafrost carbon is difficult to identify in sediments due to the contribution of fossil carbon from coal. Therefore, molecular-level techniques are required.

Here, we use molecular-level carbon isotope analysis of lipids to study permafrost turnover and export in a river catchment and fjord system on Svalbard. Our results show that lipid turnover in permafrost soils is significantly slower than in other environments, likely as a result of the low mean annual temperature and precipitation. Moreover, our results imply erosion and reburial of substantial amounts of deep permafrost soil in river and fjord sediments although the sedimentary permafrost signal is spatially heterogeneous. This spatial variability may be caused by recent soil temperature change or result from sedimentological processes.

1. Introduction

Permafrost soils (perennially frozen ground) represent a huge reservoir of organic carbon (OC) storing approximately twice the amount of C found in the present-day atmosphere and accounting for roughly 50% of the global belowground OC reservoir (Hugelius et al., 2014; Tarnocai et al., 2009). In the deeper and permanently frozen permafrost layers (below the active layer thawing during summer

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time), this OC is nearly inaccessible to microbial decomposition (Waldrop et al., 2010). However, circum-Arctic permafrost areas have experienced substantial warming during the last decades (Biskaborn et al., 2019; Christiansen et al., 2010; Romanovsky et al., 2010; S. L. Smith et al., 2010) causing permafrost thaw. Upon thaw, the OM stored in permafrost is susceptible to *in situ* microbial decomposition and enhanced mass erosion into aquatic systems including high-Arctic rivers and the Arctic Ocean (IPCC, 2013). While these aquatic systems may be characterized by even stronger microbial and photo-oxidative mineralization (Cory et al., 2013; Vonk et al., 2013), quick export of permafrost OM into marine sediments might act as efficient long-term CO₂ sink (Hilton et al., 2015). In particular, fjords in high latitudes have recently been shown to be globally important OC sinks, which may act as significant sequestration and reburial environments of mobilized permafrost OM (Cui et al., 2016; R. W. Smith et al., 2015).

Due to its particular vulnerability to warming, the high-Arctic area is of prime interest to study the quality of permafrost OC and to trace its export into fjords. Yet, substantial data gaps persist in permafrost distribution and carbon stocks for the high-Arctic region (Hugelius et al., 2014) including the Svalbard archipelago in the high European Arctic. Svalbard has experienced an unprecedented increase (ca. 2°C–3°C) of overall mean annual air temperatures during the 20th century (D'Andrea et al., 2012; Førland et al., 2011; Humlum et al., 2003; Maturilli & Kayser, 2016) and rising permafrost ground temperatures with deepening of the active layer since the 1990s (Boike et al., 2018; Christiansen et al., 2010; Isaksen et al., 2007). Accordingly, assessing OC turnover in Svalbard permafrost and characterizing and quantifying permafrost OC export into Svalbard fjords is a crucial baseline parameter to determine a potential acceleration of permafrost OC degradation/turnover and export in response to changing temperatures under Arctic amplification (Koven et al., 2015; Schädel et al., 2013, 2016; Schuur et al., 2009).

Dual carbon isotope (¹³C and ¹⁴C) analysis has been proven a powerful tool to study the quality and accessibility of permafrost OC (Dutta et al., 2006; Zimov et al., 2006) as well as the deposition of permafrost-derived OC in aquatic systems such as marine sediments (Winterfeld et al., 2015). The advantage of using a dual isotope approach lies in its ability to differentiate permafrost OC from other sources of OC such as marine production and/or to differentiate different permafrost deposit types (Dutta et al., 2006; Vonk et al., 2012; Winterfeld et al., 2015). However, the application of bulk ¹⁴C is severely limited in environments influenced by contributions from fossil (¹⁴C-free) OC sources such as kerogen or coal (Drenzek et al., 2007). On Svalbard, several coal-bearing strata primarily from the Carboniferous and Tertiary are exposed across the Spitsbergen island (Harland et al., 1976; Orvin, 1934). Accordingly, bulk $\Delta^{14}\text{C}$ values do not allow assessing OC turnover in permafrost soils and the multitude of endmembers contributing to bulk $\Delta^{14}\text{C}$ values in fjord sediments severely limits the characterization and quantification of reburied permafrost OC as it requires additional constraints to resolve mass balance calculations (e.g., Kim et al., 2011). To overcome these challenges, compound-specific radiocarbon analysis (CSRA) of vascular plant biomarkers such as alkanolic acids can be used. Long-chain alkanolic acids derive from the protective epicuticular wax cover of leaves (Eglinton & Hamilton, 1967) and they are commonly used as molecular evidence/proxies for terrestrial plants (for an overview see Diefendorf & Freimuth, 2017). CSRA of alkanolic acids has aided in understanding soil OC turnover (van der Voort et al., 2017) and has been successfully used to estimate mean residence times on the continent prior to delivery into aquatic settings (Drenzek et al., 2007; Kusch et al., 2010). In the Arctic, CSRA also has revealed the increased release and mobilization of permafrost OC in response to recent and deglacial warming (Feng et al., 2013; Meyer et al., 2019; Winterfeld et al., 2018). Yet, this tool has thus far not been used to study permafrost turnover (determined by the balance of preservation vs. degradation), in fact, only few studies to date report compound-specific alkanolic acid ¹⁴C data from soils (Matsumoto et al., 2007; van der Voort et al., 2017). Moreover, compound-specific ¹⁴C data along the land-to-ocean export continuum are as of yet missing from the high-Arctic (>70°N).

To study the quality and export of permafrost OC into a Svalbard fjord, we obtained compound-specific $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ data from permafrost soils in the Bayelva River catchment as well as sediments from the Bayelva River and Kongsfjord, a well-studied natural laboratory. We use our compound-specific alkanolic acid isotope data to determine permafrost OC turnover in the Bayelva catchment, assess export mechanisms along the flow path, and characterize the spatial dispersal and reburial of permafrost OC in Kongsfjord sediments.

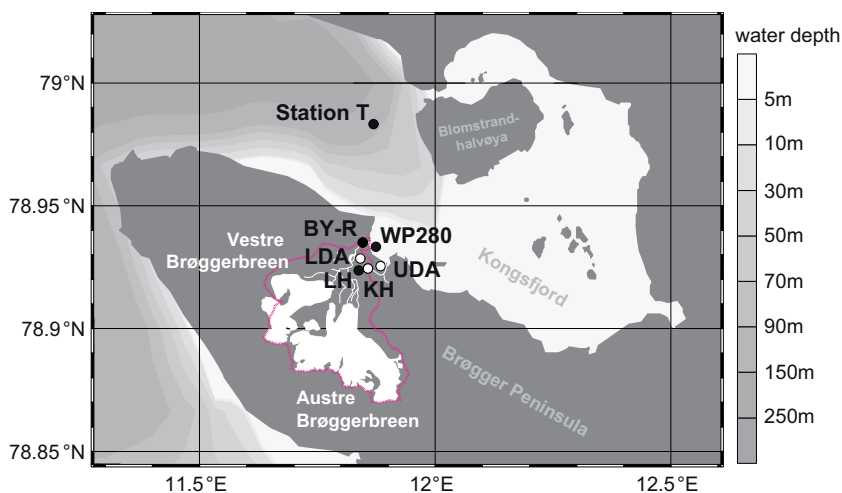


Figure 1. Study area with sampling locations. Pink dashed contour outlines the Bayelva catchment.

2. Study Area

Spitsbergen is the largest island of the Svalbard archipelago, Arctic Ocean. Its western coast is characterized by various large glacial fjord systems including the Kongsfjord. The Kongsfjord covers an area of approximately 231 km² with a maximum depth of 400 m. Several sub basins characterize the bathymetry of the fjord and since it does not have a sill at the fjord mouth restricting water mass exchange, the water column is permanently oxic (Blinova et al., 2012; Howe et al., 2003; H. Svendsen et al., 2002). Surface water derived from summer glacial freshwater run-off, ice calving, snowmelt, and precipitation overlies cold Arctic Water supplied from the East Spitsbergen Current. Underneath, a transitional/mixed water layer referred to as the transformed Arctic Water separates the Arctic water body from warmer Atlantic bottom water supplied from the West Spitsbergen Current (Nilsen et al., 2008; Rasmussen et al., 2013; H. Svendsen et al., 2002). The water column stratification is very stable during summer when the fjord is ice-free, but waters are well mixed during winters when the fjord is covered by ice. Historically, sea ice lasted approximately 7–9 months per year, but more recently the inflow of Atlantic water has prevented sea ice formation or reduced its spatial extent and thickness (Cottier et al., 2007; Pavlova et al., 2019).

The Bayelva River, Ny-Ålesund area, Brøgger Peninsula (Figure 1), drains an approximately 32 km² large catchment into the Kongsfjord. More than 50% of its catchment is glaciated by the West and East Brøgger glaciers (Vestre and Austre Brøggerbreen) and runoff is limited to June–September at the time of maximum ice melt and permafrost thaw depth (Nowak & Hodson, 2013). The river flows on a bedrock of moraine and terminates in a sandur consisting of boulders and gravel (Bogen & Bønsnes, 2003). The Bayelva catchment is underlain by permafrost with active layer depths of ~0.5–1.5 m (Boike et al., 2018; Nowak & Hodson, 2013). These permafrost soils develop on sedimentary rocks from the Late Paleozoic (conglomerates, sandstones, carbonate rocks, cherts, and siliceous limestones) as well as the Cenozoic (conglomerates, sandstones, and shales), the latter characterized by interbedded coal seams that were mined from 1916 to 1962 (Orvin, 1934; H. Svendsen et al., 2002). Pedogenesis is weak and soils are characterized primarily as haplorthels with high lithic and low nutrient content (Wojcik et al., 2019). The vegetation growing on permafrost in the Ny-Ålesund area, a polar semi-desert, during summer is dominated by bryophytes (mosses and lichens), which account for approximately 50% of the total surface area. Vascular plants follow a successional pattern with distance from the glacier and cover up to ~20%–30% of the total surface area. The most prominent vascular plant species include *Saxifraga oppositifolia*, *Dryas octopetala*, *Salix polaris*, and *Luzula confusa* while *Drepanocladus* spp. and *Cetraria delisei* are the most abundant moss and lichen species, respectively (Brossard & Joly, 1994; LLoyd, 2001; Muraoka et al., 2008). The growing period of vascular plants is short, lasting from June to August, while mosses and lichens persist from spring until autumn (LLoyd, 2001).

Table 1
Sample Locations

Station	Sample name	Latitude (°N)	Longitude (°E)	Elevation (m)	Sampling year
LH	Leirhaugen soil	78.92377	11.83810	38.0	2007
KH	Kolhaugen soil	78.92447	11.85750	45.0	2007
UDA	Upper Drainage Area soil	78.92550	11.88416	33.0	2007
LDA	Lower Drainage Area soil	78.92861	11.84138	14.0	2007
BY-R	Bayelva River	78.93433	11.84600	sea level	2008
WP280	Bayelva river mouth	78.93333	11.86950	−3.6	2008
Station T	central Kongsfjord	78.98330	11.86920	−330.0	2008

Abbreviations; BY-R, Bayelva River; KH, Kolhaugen; LH, Leirhaugen; LDA, Lower Drainage Area; UDA, Upper Drainage Area

3. Materials and Methods

3.1. Materials

The sampling strategy was designed to obtain an export transect sample set covering the permafrost soil-river-ocean system. Samples were taken during several land- and ship-based campaigns. In 2007, soil samples from the Bayelva River catchment were collected from the thawed active layer in the northern foreland of the East Brøgger glacier (Figure 1, Table 1). Leirhaugen (LH) is adjacent to the main stem of the Bayelva River (Boike et al., 2008) and permafrost ground temperatures have been monitored at this site since 1998 (Boike et al., 2018; Roth & Boike, 2001). Additional soil profiles in the catchment were sampled from Kolhaugen (KH), Upper Drainage Area (UDA), and Lower Drainage Area (LDA) as described by Rethemeyer et al. (2010). Briefly, soil pits (ca. 1.5 m²) were opened to up to ~1 m depth (Tables S1 and S2) at each site, reaching either the permafrost table or bedrock-like ground that precluded further excavation. All profiles were sub-sampled based on visible redox characteristics (e.g., color), typically representing three mineral soil horizons. Soil samples were stored frozen (−20°C) in precombusted glass jars until analysis. A sample of coal was also obtained from the catchment.

Sediment samples from the Bayelva River (BY-R), its river mouth (WP280), and the central Kongsfjord (Station T) were obtained in 2008 using a shovel (BY-R), push cores operated by divers (WP280; 10 cm total sediment recovery) or a hand-operated HAPS corer (Station T; 14 cm total sediment recovery). All sediment samples were sliced into 0–1 or 0–2 cm segments and stored in precombusted glass jars at −20°C until analysis.

3.2. Methods

Soil samples were sieved through a 2 mm sieve and visible coal particles were removed. Soil and sediment samples were freeze-dried, homogenized, and Soxhlet-extracted for 48 h (following a 24 h cellulose thimble pre-extraction) using a 90:10 dichloromethane: methanol (v:v) solvent mixture (Kusch et al., 2016). The total lipid extract was processed and alkanolic acids were separated from neutral lipids, purified, and converted to fatty acid methyl esters (FAMES) according to the methods described in Mollenhauer and Eglinton (2007), except AgNO₃ column chromatography was performed instead of urea adduction. A split of the fatty acid methyl ester fraction was taken for routine quantitative flame-ionization gas chromatography (GC-FID) analysis, and concentrations were calculated using an external FAME standard mixture. For isolation of FAME homologs, preparative capillary gas chromatography (PCGC) was used according to the method described in Kusch et al. (2010). To remove potential contamination from column bleed, FAMES were eluted over a silica gel column after isolation using hexane: dichloromethane (2:1 v:v). This PCGC isolation procedure was checked for blank carbon contribution at the time and no significant contamination was detected (Mollenhauer & Rethemeyer, 2009). Based on individual FAME quantities, homologs were pooled and samples were converted to CO₂. FAMES were transferred into precombusted (900°C) quartz tubes and 150 µg precombusted copper oxide were added as oxygen source. The quartz

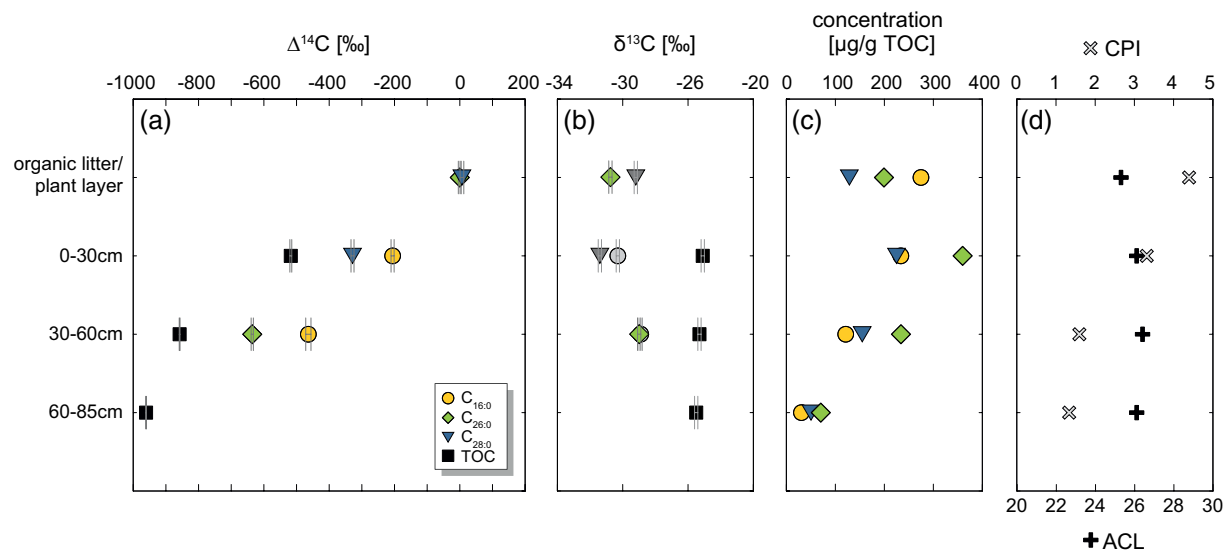


Figure 2. The $\Delta^{14}\text{C}$ (a) and $\delta^{13}\text{C}$ (b) carbon isotopic composition of bulk OC and alkanolic acids in the Leirhaugen permafrost soil profile. Corresponding concentrations (c) and CPI and ACL (d) of individual alkanolic acids in the profile. Error bars show 1σ analytical uncertainty. ACL, average chain length; CPI, carbon preference index, OC, organic carbon.

tubes were evacuated while immersed in dry ice, flame-sealed, and combusted at 900°C for 4 h. Resulting CO_2 was stripped of water and quantified. Samples were ^{14}C -analyzed at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) Facility at Woods Hole Oceanographic Institution and the CologneAMS facility at the University of Cologne. AMS measurements of bulk OC were carried out using standard methods (McNichol et al., 1994; Rethemeyer et al., 2013), compound-specific AMS radiocarbon measurements were performed following dedicated techniques for small sample sizes (Pearson et al., 1998). Splits of the CO_2 gas samples were used to measure the stable carbon isotopic composition on a VG Optima IRMS. Radiocarbon results are reported as conventional ^{14}C ages in years BP and $\Delta^{14}\text{C}$ in ‰ according to Stuiver and Polach (1977), including correction for kinetic fractionation based on $\delta^{13}\text{C}$ values. The $\delta^{13}\text{C}$ values are reported in ‰ relative to VPDB. Both $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ values of individual alkanolic acids were corrected for the addition of one methyl group during derivatization using isotope mass balance.

4. Results

4.1. Elemental and Molecular Geochemical Data

Organic carbon contents (%OC) in the permafrost soil profiles range from 0.1% to 3.3% (Tables S1 and S2) with the lowest %OC typically observed in the deepest sample of each profile. The organic litter/plant layers have %OC values from 19.9% to 28.4%. Alkanolic acid concentrations in the permafrost soils and organic litter/plant layers show a bimodal distribution with short-chain maxima centering on $\text{C}_{16:0}$ or $\text{C}_{18:0}$ and long-chain maxima centering on $\text{C}_{24:0}$ or $\text{C}_{26:0}$. The average chain length (ACL: $[\sum n \times \text{C}_n]/[\sum \text{C}_n]$; $n = 16\text{--}32$) ranges from 25.3 to 26.4 in the LH profile, 22.7 to 25.0 in the KH trough profile, 24.1 to 25.4 in the KH center profile, 24.7 to 26.3 in the UDA profile, and 25.2 to 27.4 in the LDA profile, respectively. ACL distributions (Figures 2 and S1) do not show a common trend between permafrost profiles. Carbon preference index (CPI: $0.5 \times [\sum \text{C}_{20\text{--}30}/\sum \text{C}_{19\text{--}29} + \sum \text{C}_{20\text{--}30}/\sum \text{C}_{21\text{--}31}]$) values range from 4.4 to 1.4 (LH), 4.4 to 1.5 (KH trough), 1.6 to 2.2 (KH center), 7.0 to 1.8 (UDA), and 3.7 to 3.5 (LDA). Overall, CPI values decrease with depth throughout the profile, but some profiles show a slight increase in the lowermost depth (Figures 2 and S1). CPI values in the mineral soils are consistent with values determined for bacterial biomass (Wiesenberg et al., 2012). Unfortunately, alkanolic acid concentrations in the KH, UDA, and LDA soil profiles (Table S2) were one to two orders of magnitude lower (per gram dry weight) than in LH, thus, restricting compound-specific ^{14}C analyses to LH.

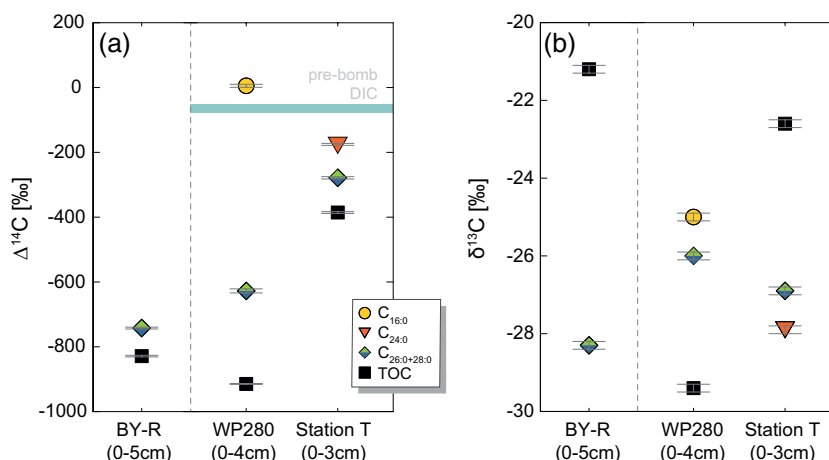


Figure 3. The $\Delta^{14}\text{C}$ (a) and $\delta^{13}\text{C}$ (b) carbon isotopic composition of bulk OC and alkanolic acids in the Bayelva River and Kongsfjord surface sediments. Note that the pooled long-chain alkanolic acid sample at station WP280 also includes $\text{C}_{24:0}$ alkanolic acid and was obtained from 4–6 cm depth. Error bars show 1σ analytical uncertainty. “Pre-bomb DIC” denotes the $\Delta^{14}\text{C}$ range of DIC determined for Spitsbergen fjords by Mangerud and Gulliksen (1975). DIC, dissolved inorganic carbon; OC, organic carbon.

Sedimentary %OC ranges from 0.1% (BY-R) to 3.6% (Station T; Tables S1 and S2). Alkanolic acid distributions are also bimodal in the sediments, but short-chain alkanolic acids are significantly more abundant (up to 2 orders of magnitude) than long-chain alkanolic acids in cores WP280 and Station T whereas they are present in similar abundance in BY-R. The high contribution of short-chain alkanolic acids is mirrored in the corresponding ACL values that range from 16.8 (Station T) to 20.9 (WP280 6–8 cm) in the fjord sediments and is 24.9 in sample BY-R. CPI values range from 1.6 (BY-R) to 2.5 (WP280 0–2 cm).

4.2. Bulk OC Isotope Data

Marine surface sediment (0–2 cm) OC $\Delta^{14}\text{C}$ data (Figure 3, Table 2) range from $-385.7 \pm 2.7\text{‰}$ (Station T) to $-914.6 \pm 1.0\text{‰}$ (WP280). OC $\Delta^{14}\text{C}$ values at station WP280 decrease further downcore from $-914.6 \pm 1.0\text{‰}$ (0–2 cm) to $-985.1 \pm 1.0\text{‰}$ (8–10 cm). Similarly, the $\Delta^{14}\text{C}$ values from the LH soil profile (Figure 2) decrease with depth from $-517.5 \pm 2.9\text{‰}$ (LH 0–30 cm) to $-960.8 \pm 0.7\text{‰}$ (LH 60–85 cm). OC from the Bayelva River (BY-R) reflects a $\Delta^{14}\text{C}$ value of $-828.7 \pm 2.4\text{‰}$.

OC $\delta^{13}\text{C}$ values range from $-25.1 \pm 1.0\text{‰}$ (0–30 cm) to $-25.5 \pm 1.0\text{‰}$ (60–85 cm) in the LH soil profile and are $-21.2 \pm 1.0\text{‰}$ in the river sediment (BY-R) and $-22.6 \pm 1.0\text{‰}$ at Station T.

4.3. Alkanolic Acid Isotope Data

The $\Delta^{14}\text{C}$ value (Figure 3, Table 2) of short-chain $\text{C}_{16:0}$ alkanolic acid at station WP280 is $-73.3 \pm 3.3\text{‰}$. For the LH soil profile $\text{C}_{16:0}$ alkanolic acid $\Delta^{14}\text{C}$ values (Figure 2) are significantly more depleted ranging from $-205.9 \pm 4.7\text{‰}$ (LH 0–30 cm) to $-463.8 \pm 8.0\text{‰}$ (LH 30–60 cm). Long-chain $\text{C}_{26:0}$ and $\text{C}_{28:0}$ (or combined $\text{C}_{26:0+28:0}$) alkanolic acid $\Delta^{14}\text{C}$ values range from $-278.6 \pm 3.6\text{‰}$ ($\text{C}_{26:0+28:0}$; Station T) to $-627.5 \pm 6.2\text{‰}$ ($\text{C}_{26:0+28:0}$; WP280 4–6 cm). Unfortunately, low alkanolic acid concentrations at 60–85 cm depth precluded CSRA. At all stations where multiple alkanolic acid $\Delta^{14}\text{C}$ values are available, $\Delta^{14}\text{C}$ values are more depleted with increasing chain length except for $\text{C}_{26:0}$ and $\text{C}_{28:0}$ in the organic litter/plant layer of the LH soil profile, which agree within 1σ analytical uncertainty.

High molecular weight $\text{C}_{26:0}$ and $\text{C}_{28:0}$ alkanolic acid $\delta^{13}\text{C}$ values range from $-29.0 \pm 0.1\text{‰}$ (30–60 cm) to $-31.4 \pm 0.1\text{‰}$ (0–30 cm) in the soil profile and the $\delta^{13}\text{C}$ value of $\text{C}_{26:0+28:0}$ alkanolic acids in the Bayelva sediment is $-28.3 \pm 0.1\text{‰}$. In the fjord, HMW alkanolic acid $\delta^{13}\text{C}$ values range from $-26.0 \pm 0.1\text{‰}$ ($\text{C}_{24:0+26:0+28:0}$; WP280 4–6 cm) to $-27.9 \pm 0.1\text{‰}$ ($\text{C}_{24:0}$; Station T). The $\delta^{13}\text{C}$ values of LMW $\text{C}_{16:0}$ alkanolic acid in the LH soil profile are $-30.3 \pm 0.1\text{‰}$ at 0–30 cm depth and $-28.9 \pm 0.1\text{‰}$ at 30–60 cm depth (Table 2) and $-25.1 \pm 0.1\text{‰}$ in the Kongsfjord surface sediment. The $\text{C}_{24:0}$ alkanolic acid $\delta^{13}\text{C}$ value at Station T is $-27.9 \pm 0.1\text{‰}$.

Table 2

Radiocarbon and Stable Carbon Isotopic Composition ($\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ Given in ‰) of Soil (Leirhaugen), River (Bayelva) and Fjord (Kongsfjord) sediments. Errors Given as 1σ Analytical Uncertainty (Propagated for Alkanoic acids)

Sample	$\Delta^{14}\text{C}$ (‰)	^{14}C age (years BP)	$\delta^{13}\text{C}$ (‰)	AMS ID No.
<i>Leirhaugen</i>				
LH (organic litter/plant layer)				
C _{26:0} alkanoic acid	-0.2 ± 4.8	bomb- ^{14}C	-30.7 ± 0.1	NOSAMS 74182
C _{28:0} alkanoic acid	6.4 ± 6.0	bomb- ^{14}C	-29.2 ± 0.1	NOSAMS 74183
LH (0–30 cm)				
C _{16:0} alkanoic acid	-205.9 ± 5.0	$1,800 \pm 50$	-30.3 ± 0.1	NOSAMS 74184
C _{28:0} alkanoic acid	-328.7 ± 4.9	$3,140 \pm 60$	-31.4 ± 0.1	NOSAMS 74185
bulk OC	-517.5 ± 2.9	$5,800 \pm 50$	-25.1 ± 0.1	NOSAMS 66572
LH (30–60 cm)				
C _{16:0} alkanoic acid	-463.8 ± 8.2	$4,950 \pm 120$	-28.9 ± 0.1	NOSAMS 74186
C _{26:0} alkanoic acid	-635.5 ± 3.8	$8,050 \pm 85$	-29.0 ± 0.1	NOSAMS 74187
bulk OC	-858.0 ± 1.3	$15,600 \pm 70$	-25.3 ± 0.1	NOSAMS 66573
LH (60–85 cm)				
bulk OC	-960.8 ± 0.7	$26,000 \pm 130$	-25.5 ± 0.1	NOSAMS 66574
<i>Bayelva River</i>				
BY-R (0–5 cm)				
C _{26:0+28:0} alkanoic acid	-742.1 ± 3.3	$10,830 \pm 100$	-28.3 ± 0.1	NOSAMS 74188
bulk OC	-828.7 ± 2.4	$14,100 \pm 110$	-21.2 ± 0.1	NOSAMS 66576
<i>Kongsfjord</i>				
WP280 (0–4 cm)				
C _{16:0} alkanoic acid ^a	10.4 ± 5.6	bomb- ^{14}C	-25.0 ± 0.1	NOSAMS 74189
C _{16:0} alkanoic acid ^a	0.1 ± 4.2	bomb- ^{14}C	-25.0 ± 0.1	NOSAMS 74190
bulk OC (0–2 cm)	-914.6 ± 1.0	$19,750 \pm 130$	n.d.	COL1543.1.2
WP280 (4–6 cm)				
C _{24:0+26:0+28:0} alkanoic acid	-628.0 ± 6.5	$7,890 \pm 140$	-26.0 ± 0.1	NOSAMS 74191
bulk OC	-981.1 ± 1.0	$31,750 \pm 530$	n.d.	COL1544.1.2
WP280 (8–10 cm)				
bulk OC	-985.1 ± 1.0	$33,700 \pm 500$	n.d.	COL1545.1.1
Station T (0–3 cm)				
C _{24:0} alkanoic acid	-175.4 ± 3.5	$1,490 \pm 35$	-27.9 ± 0.1	NOSAMS 89091
C _{26:0+28:0} alkanoic acid	-278.6 ± 4.0	$2,560 \pm 45$	-26.9 ± 0.1	NOSAMS 89092
bulk OC	-385.7 ± 2.7	$3,850 \pm 35$	-22.6 ± 0.1	NOSAMS 83355

Errors given as 1σ analytical uncertainty (propagated for alkanoic acids).

Abbreviations: BY-R, Bayelva River; LH, Leirhaugen; n.d., not determined; OC, organic carbon.

^aSample was split prior to AMS measurements.

5. Discussion

5.1. Isotope Source Signatures of Permafrost OC in the Bayelva Catchment

The LH soil profile is characterized by bulk OC $\Delta^{14}\text{C}$ values that are significantly depleted and decrease with depth (Figure 2, Table S2), a pattern that unlikely reflects long-term radioactive decay of biomass alone, but rather results from the contribution of coal particles to the bulk OC pool (Kim et al., 2011). This interpretation may be supported by the very good agreement of the LH bulk OC $\delta^{13}\text{C}$ values (mean -25.3‰) with

the $\delta^{13}\text{C}$ value of coal (mean -25.0‰) from the Bayelva catchment analyzed by Kim et al. (2011), although coal-free bulk soil OC or lichen may have similar $\delta^{13}\text{C}$ isotopic compositions (Zwolicki et al., 2016). Concurrent compound-specific $\Delta^{14}\text{C}$ values of both short-chain $\text{C}_{16:0}$ and long-chain $\text{C}_{26:0}$ and $\text{C}_{28:0}$ alkanolic acids are significantly higher than bulk OC values (confirming coal contributions to bulk OC) but also decrease from the organic litter/plant layer into deeper mineral soil horizons (Figure 2). Rethemeyer et al. (2010) demonstrated that these alkanolic acids derive mainly from bacteria, moss, lichen, and tundra vegetation. However, alkanolic acids have previously also been detected in coal (Baset et al., 1980; Niwa et al., 1988; Snape et al., 1981). The LH alkanolic acid inventories may, thus, contain a coal-derived subpool that may potentially bias $\Delta^{14}\text{C}$ values although alkanolic acids have not been reported from coal on Svalbard (Ćmiel & Fabiańska, 2004; Marshall et al., 2015). While we could detect alkanolic acids in the coal sample from the catchment, multiple lines of evidence suggest that the alkanolic acid signature is not authigenic but rather stems from intrusion of modern OC: The overall alkanolic acid concentrations in the coal were much lower than observed in the soil profiles (especially for long-chain homologs) and $\text{C}_{16:1}$ and $\text{C}_{18:1}$ alkanolic acids had the highest individual concentrations (Table S2). Short-chain monoenoic alkanolic acids degrade during early coalification (Řezanka, 1992) and typically have a microbial origin (Zelles, 1999). Moreover, Svalbard coals are strongly coalified as indicated by high maceral content and high vitrinite reflection classifying them as bituminous coal (Ćmiel & Fabiańska, 2004; Marshall et al., 2015), which commonly only show traces of long-chain alkanolic acids if any (Baset et al., 1980; Niwa et al., 1988; Řezanka, 1992; Snape et al., 1981). In addition, individual $\delta^{13}\text{C}$ values of alkanolic acids in the LH profile agree with those in the organic litter/plant layer (Figure 2, Table S2), which represents recently decomposed vegetation as confirmed by the bomb- ^{14}C signature of litter/plant layer $\text{C}_{26:0}$ and $\text{C}_{28:0}$ alkanolic acid. Even if coal-derived long-chain alkanolic acids contributed to the LH alkanolic acid inventories, their influence on the alkanolic acid $\Delta^{14}\text{C}$ values should be negligible (as per mass balance) given that the LH alkanolic acid abundances are 1–2 orders of magnitude higher than those observed in the coal sample. Accordingly, the strong ^{14}C -depletion ($-205.9 \pm 5.0\text{‰}$ to $-635.5 \pm 3.8\text{‰}$) of alkanolic acids in the LH mineral soil with respect to the atmosphere ($\sim 50\text{‰}$) at the time of sampling in 2007 (Levin et al., 2013) implies slow degradation and radioactive decay of biomass, likely due to freeze-locking and less favorable conditions for microorganisms at greater soil depth.

5.2. Permafrost Turnover in the Bayelva Catchment Inferred from Alkanolic Acids

The decrease of alkanolic acid $\Delta^{14}\text{C}$ values with depth mirrors globally observed bulk $\Delta^{14}\text{C}$ patterns that are attributed to result from the complex interaction of aboveground input, organic matter turnover, and vertical mixing through bioturbation, leaching, and erosion (Mathieu et al., 2015; Shi et al., 2020). Since the permafrost soils in the Bayelva catchment are mostly frozen throughout the year and active layer thaw is limited to the peak summer months, microbial turnover likely dominates these processes since vegetation is sparse and bioturbation is virtually absent. Instead, cryoturbation may lead to OC mixing in permafrost soil (Bockheim, 2007). However, all investigated soil profiles in the Bayelva catchment (LH, KH, UDA, and LDA) indicate little distortion by cryoturbation since bulk OC content (Table S1), alkanolic acid concentrations (Figures 2, and S1), and bacterial intact polar lipid and bacteriohopanepolyol concentrations (Rethemeyer et al., 2010) decrease with depth in all profiles rather than showing more random distributions (Bockheim, 2007). This observation is in good agreement with a recent study by Wojcik et al. (2019) who also found little evidence for cryoturbation on the Brøgger Peninsula. Accordingly, the decreasing alkanolic acid $\Delta^{14}\text{C}$ values in the LH profile and of OC elsewhere on Svalbard (Cherkinsky, 1996) are consistent with a primary control of freeze-locking and degradation/radioactive decay on OC inventories and $\Delta^{14}\text{C}$ values in the Bayelva catchment. Since we cannot unequivocally determine the contribution of coal to bulk OC and its influence on $\Delta^{14}\text{C}$ values, we will use the alkanolic acid $\Delta^{14}\text{C}$ values to assess turnover in the LH soil and by inference, the Bayelva catchment (assuming the LH biomarker distributions are representative for the catchment, which is supported by the good agreement of biomarker abundances with the KH, UDA, and LDA profiles).

The $\text{C}_{16:0}$, $\text{C}_{26:0}$, and $\text{C}_{28:0}$ alkanolic acid $\Delta^{14}\text{C}$ values from 0–30 cm depth or 30–60 cm depth ($-205.9 \pm 5.0\text{‰}$ to $-635.5 \pm 3.8\text{‰}$) correspond to conventional ^{14}C ages of $1,800 \pm 50$ to $8,050 \pm 85$ years BP (Table 2, Figure 4a) implying very slow (multi-millennial) alkanolic acid turnover in the LH mineral soil. Here, we will assess alkanolic acid turnover in the LH mineral soils, a function of input/preservation and output/degradation,

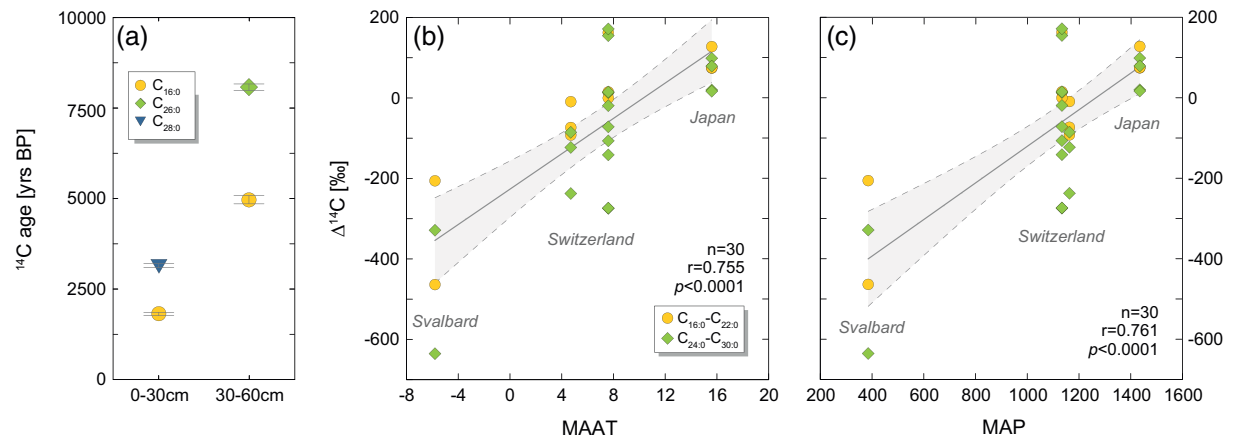


Figure 4. Alkanoic acid radiocarbon ages in the Leirhaugen soil profile (a). Scatter plots of (b) mean annual air temperature (MAAT) and (c) mean annual precipitation (MAP) against the $\Delta^{14}\text{C}$ carbon isotopic composition of short-chain and long-chain alkanoic acids (even chain lengths only) in soils from Svalbard (this study), Switzerland (from van der Voort et al., 2017; the subalpine site is characterized by lower MAAT and slightly higher MAP than the temperate site), and Japan (from Matsumoto et al., 2007). In panels (b) and (c), alkanoic acids were grouped according to chain length although data points represent individual homologs except for the pooled $\text{C}_{16:0-22:0}$ alkanoic acid samples from Swiss soils; error bars are smaller than symbol size and are omitted for better visibility.

using ^{14}C ages and $\Delta^{14}\text{C}$ values rather than calculating turnover times following the approach outlined, for example, by Torn et al. (2009) since the results cannot be reconciled with the environmental history. Turnover time can be calculated assuming either steady state (input flux equals output flux) or open system behavior. While subzero temperatures retain the soils in a state with very limited input and output fluxes throughout most of the year, pedogenesis is likely on-going on the Brøgger Peninsula. However, modeling turnover times for LH with open system behavior (and absence of bomb- ^{14}C) requires a soil formation age in considerable excess of the $\sim 10,000$ years since deglaciation and emergence of the Brøgger Peninsula (Forman et al., 1987). Modeling alkanoic acid turnover times assuming steady state (i.e., turnover time equals the inverse of the decomposition rate) yields turnover times as high as $14,410 \pm 240$ years for $\text{C}_{26:0}$ alkanoic acid at 30–60 cm depth, thus, also exceeding the Holocene framework for soil development and implying that soils are indeed not at steady state.

Turnover decreases both with depth in the profile and with chain length. In the mineral soil horizons, $\text{C}_{16:0}$ alkanoic acid ^{14}C ages increase from $1,800 \pm 50$ to $4,950 \pm 120$ and $\text{C}_{26:0}$ or $\text{C}_{28:0}$ alkanoic acid ^{14}C ages increase from $3,140 \pm 60$ to $8,050 \pm 85$ from 0–30 cm to 30–60 cm, respectively (Figure 4a). This pattern has previously been observed in soils by van der Voort et al. (2017) and Matsumoto et al. (2007), the only studies to date reporting compound-specific alkanoic acid ^{14}C data for individual homologs from soils, and may be the result of different sources or different turnover/recalcitrance of individual homologs, or a combination thereof. In soils, long-chain alkanoic acids derive from vascular plants and represent a slow cycling alkanoic acid pool, whereas both vascular plants and microbes are synthesizers (or recyclers) of $\text{C}_{16:0}$ alkanoic acid that represents a relatively fast cycling pool (van der Voort et al., 2017), an observation that is confirmed by the LH data. On the Brøgger Peninsula, the organic layer and uppermost 30 cm of soils together store $\sim 80\%$ of the soil OC (Wojcik et al., 2019). Accordingly, the lower turnover of alkanoic acids at depth should primarily result from strongly limited vertical transport of modern OC into the deeper mineral soil and very slow degradation/decomposition rates. In addition, the even shorter thaw period of the deep active layer may further limit/slow down microbial degradation.

While compound-specific $\Delta^{14}\text{C}$ values have rarely been determined for soils, the limited information available indicates that alkanoic acid turnover is much faster in non-Arctic regions, analogous to global soil bulk $\Delta^{14}\text{C}$ (Shi et al., 2020). Alkanoic acids in one soil sample from Sapporo, Japan all contain bomb- ^{14}C indicating decadal turnover (Matsumoto et al., 2007). Short-chain and long-chain alkanoic acids from a temperate soil profile in Switzerland range from $171.5 \pm 0.6\text{‰}$ to $-273.7 \pm 5.8\text{‰}$ (van der Voort et al., 2017) with the majority of $\Delta^{14}\text{C}$ values containing bomb- ^{14}C which implies primarily decadal turnover. In the same study, alkanoic acid $\Delta^{14}\text{C}$ values at a subalpine Swiss site range from $-9.2 \pm 1.2\text{‰}$ to $-274.4 \pm 0.4\text{‰}$ (van der Voort

et al., 2017) indicating centennial to millennial turnover. In comparison with the alkanolic acid $\Delta^{14}\text{C}$ values determined for both Swiss locations, the ^{14}C -depletion for the fast ($\text{C}_{16:0}$ alkanolic acid) and slow ($\text{C}_{26:0}$ and $\text{C}_{28:0}$ alkanolic acid) cycling alkanolic acid pools in each soil horizon and between soil horizons is significantly higher in the LH profile.

Globally, both temperature and precipitation are considered the primary factors influencing soil OC turnover, with different relative contributions in different climate regimes (Carvalhais et al., 2014; Chen et al., 2013). Comparison of the alkanolic acid $\Delta^{14}\text{C}$ data from Svalbard, Switzerland, and Japan soils with climatic data shows that while the $\Delta^{14}\text{C}$ amplitude is quite high at each site depending on alkanolic acid chain length and soil depth, alkanolic acid $\Delta^{14}\text{C}$ values decrease with both mean annual air temperature and mean annual precipitation (Figures 4b and 4c). Accordingly, alkanolic acid turnover, that is, output/degradation, increases with temperature and precipitation, in agreement with the global soil OC observations. Given the high vulnerability of the high-Arctic to warming (IPCC, 2013), alkanolic acid/permafrost turnover/degradation may likely accelerate in the Bayelva catchment in the future. Deepening of the active layer has been observed on Svalbard since the 1990s (Christiansen et al., 2010; Isaksen et al., 2007) and daily temperature data from 1998 to 2017 show a 3°C – 4°C increase in mean annual soil temperature of the active layer (up to 60 cm depth) and permafrost (ca. 1.4 m depth) on Leirhaugen (Boike et al., 2018). Warmer soil temperatures and an extended summer thaw period will likely promote enhanced microbial activity and turnover.

5.3. Tracing Permafrost OC Export into Sediments

5.3.1. Permafrost OC in the Bayelva River

The seasonally active Bayelva River is characterized by high runoff and sediment load, and bank erosion and sliding are common geomorphological processes along its flow path that likely account for the majority of permafrost export (Bogen & Bønsnes, 2003; Nowak & Hodson, 2014). Fine sediment accumulation is patchy in the Bayelva and primarily derives from the suspended load transported during snowmelt and rain floods (Bogen & Bønsnes, 2003), thus, the BY-R sediment sample represents recently eroded/mobilized material. Bulk OC is strongly ^{14}C -depleted ($-828.7 \pm 2.4\text{‰}$) and analogous to the LH samples, likely biased by the contribution of coal particles (Figure 3). Notably, the bulk OC $\delta^{13}\text{C}$ value in sample Bayelva River surface sediment sample (BY-R) is much higher (by roughly 4‰) than observations from LH and even the coal investigated by Kim et al. (2011), while the $\text{C}_{26:0+28:0}$ alkanolic acid $\delta^{13}\text{C}$ value agrees within approximately 1‰ – 2‰ with alkanolic acid $\delta^{13}\text{C}$ values from LH. This pattern suggests another yet also strongly ^{14}C -depleted OC pool contributing to bulk OC in Bayelva River. Given the high runoff and high sediment load (Bogen & Bønsnes, 2003) as well as the ^{14}C -depletion of bulk OC, we consider autotrophic production in the river unlikely. Instead, we suspect that ice-rafted debris (IRD; mean $\delta^{13}\text{C} = -22.6\text{‰}$ and $\Delta^{14}\text{C} = -809\text{‰}$) may contribute to the sample (Kim et al., 2011).

The $\text{C}_{26:0+28:0}$ alkanolic acids in BY-R are strongly ^{14}C -depleted ($\Delta^{14}\text{C} = -742.1 \pm 3.3\text{‰}$), in fact, their $\Delta^{14}\text{C}$ value is roughly 100‰ lower than that of $\text{C}_{26:0}$ alkanolic acid at 30–60 cm depth ($\Delta^{14}\text{C} = -635.5 \pm 3.4\text{‰}$) in the LH soil profile (Figures 2 and 3). While bulk OC may contain OC from IRD, we do not expect this OC to be a major source of long-chain alkanolic acids to the Bayelva sediment. Glacier OC and runoff is typically dominated by anthropogenic hetero-aromatic aerosols with only minor contribution of saturated hydrocarbons (Grannas et al., 2006; Stubbins et al., 2012). If the Brøgger glaciers indeed contained alkanolic acids, they likely derive from erosion of basal permafrost from the catchment. Both, basal erosion as well as bank erosion of permafrost provide a mechanism for the deposition of a small sediment entity that originates from the deep active layer or permafrost and is characterized by strongly ^{14}C -depleted $\text{C}_{26:0+28:0}$ alkanolic acids. As such, the BY-R sample illustrates that ^{14}C -depleted long-chain alkanolic acids from the deep active layer and/or permafrost are effectively exported to the Bayelva River (and consequently further into the Kongsfjord), a mechanism for which direct molecular-level evidence has thus far not been reported.

5.3.2. Permafrost OC in the Kongsfjord

In the Kongsfjord, sedimentary bulk OC consists of a mixture of marine OC, permafrost OC, and fossil OC (coal and IRD) and the relative contributions of each endmember vary spatially within the fjord depending

on the distance to glaciers or rivers, respectively (e.g., Kim et al., 2011). The investigated core-top sediments are of decadal age (Supplement), thus, any ^{14}C -depletion results from supply of ^{14}C -depleted OC rather than sedimentological processes such as nondeposition or erosion. Off the Bayelva River, the core-top sediment bulk OC at station WP280 is strongly ^{14}C -depleted ($\Delta^{14}\text{C} = -914.6 \pm 1.0\text{‰}$) revealing substantial contributions of OC containing coal particles. The bulk OC ^{14}C -depletion at this particular site is significantly higher than previously observed in Kongsfjord by Kim et al. (2011), who report values ranging from -203‰ to -735‰ , and may be explained by the close proximity to the Bayelva River. At Station T, located in the central Kongsfjord, the bulk OC $\Delta^{14}\text{C}$ value is significantly higher ($-385.7 \pm 2.7\text{‰}$), reflecting enhanced contributions from marine OC and reduced input of OC from permafrost and coal. We trace marine OC in the fjord sediments using the compound-specific $\Delta^{14}\text{C}$ data from short-chain alkanolic acids at station WP280, which predominantly derive from marine phytoplankton and ice algae (Leu et al., 2006; McMahon et al., 2006; Søreide et al., 2008). Although $\text{C}_{16:0}$ alkanolic acids also occur in the LH soil samples, they are characterized by significantly higher $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values at station WP280 compared to the LH soil $\text{C}_{16:0}$ alkanolic acids. Additional support for a marine origin comes from significantly higher $\text{C}_{16:0}$ alkanolic acid concentrations and a much lower ACL in the WP280 core-top (17.4; Table S2) than observed in the LH profile (>25.3 ; Figure 2). The $\text{C}_{16:0}$ alkanolic acids off the Bayelva River are enriched in ^{14}C (mean $5.3 \pm 4.6\text{‰}$) with respect to the pre-bomb surface dissolved inorganic carbon $\Delta^{14}\text{C}$ value of $-57 \pm 6\text{‰}$ to $-67 \pm 9\text{‰}$ (Figure 3) obtained from several Spitsbergen fjords (Mangerud & Gulliksen, 1975) attesting to deposition after 1950, which is consistent with the independently derived sediment ages.

In contrast to the planktonic short-chain alkanolic acids, permafrost-derived long-chain alkanolic acids are strongly ^{14}C -depleted (Figure 3), particularly the $\text{C}_{24:0+26:0+28:0}$ alkanolic acids at station WP280 that have a $\Delta^{14}\text{C}$ value of $-628.0 \pm 6.5\text{‰}$. The $\Delta^{14}\text{C}$ values in the fjord sediments agree well with the $\Delta^{14}\text{C}$ values of $\text{C}_{28:0}$ alkanolic acid at 30–60 cm depth in the LH profile ($-635.5 \pm 3.8\text{‰}$) as well as the BY-R sample ($-742.1 \pm 3.3\text{‰}$), but unlikely results from erosion of specific permafrost depth horizons. Instead, the alkanolic acid $\Delta^{14}\text{C}$ value in the WP280 surface sediment represents the average alkanolic acid $\Delta^{14}\text{C}$ value of the eroded permafrost depth horizons weighted by their relative volumetric contributions and alkanolic acid inventories. This implies that substantial amounts of deep active layer and/or permafrost must be transported to station WP280. In contrast to $\text{C}_{24:0+26:0+28:0}$ alkanolic acids at station WP280, $\text{C}_{26:0+28:0}$ alkanolic acids at offshore Station T have a significantly higher $\Delta^{14}\text{C}$ value ($-278.6 \pm 4.0\text{‰}$). This offset may be even more pronounced if the pooled alkanolic acid sample at station WP280 is in part influenced by $\text{C}_{24:0}$ alkanolic acid of marine origin (Volkman et al., 1980, 1998), which would likely have a less ^{14}C -depleted signature similar to $\text{C}_{24:0}$ alkanolic acid at Station T (Figure 3). Several physical processes may explain the alkanolic acid $\Delta^{14}\text{C}$ offset observed at both sites. Due to the very close proximity of site WP280 to the Bayelva River, it exclusively receives terrestrial input from the Bayelva catchment. In contrast, Station T may receive input from other permafrost areas such as Blomstrandhalvøya (Figure 1) that may have different alkanolic acid $\Delta^{14}\text{C}$ signatures than the Bayelva catchment. If Station T indeed accumulates permafrost OC primarily from the Brøgger Peninsula/Bayelva catchment, transport to this site may be by associated with a grain size/sorting effect on alkanolic acid $\Delta^{14}\text{C}$ values. Previous observations show that alkanolic acid $\Delta^{14}\text{C}$ values typically increase with grain size (Bao et al., 2018; Yu et al., 2019); however, Bao et al. (2018) also found the opposite trend in Washington Margin sediments. In addition to storing an estimated $\sim 70\%$ of soil OC on the Brøgger Peninsula, the uppermost 30 cm of mineral soil are also characterized by lower fine fraction bulk density (Wojcik et al., 2019) and based on the LH profile, less ^{14}C -depleted $\text{C}_{26:0}$ alkanolic acids ($\Delta^{14}\text{C} = -328.7 \pm 4.9\text{‰}$). Thus, higher density/larger grain size material containing ^{14}C -depleted long-chain alkanolic acids from deeper mineral soils may only be deposited near station WP280 while the lower density/smaller grain size material is preferentially transported to Station T. Finally, sedimentation rates at Station T are lower than at station WP280 and the respective alkanolic acid $\Delta^{14}\text{C}$ values may to some extent also record a change in permafrost OC erosion associated with the recent soil temperature change in the Bayelva catchment (Boike et al., 2018).

Our data do not allow to unambiguously identify the process responsible for the observed $\Delta^{14}\text{C}$ offset between $\text{C}_{24:0+26:0+28:0}$ alkanolic acids at station WP280 and $\text{C}_{26:0+28:0}$ alkanolic acids at Station T. Irrespective, however, our data illustrate that even at the compound-specific level sedimentary $\Delta^{14}\text{C}$ values of permafrost-derived OC seem to be heterogenous in the Kongsfjord. Accordingly, bulk OC $\Delta^{14}\text{C}$ values may not only be influenced by varying degrees of coal contributions admixed to permafrost and marine OC, but

also a nonsteady state permafrost OC $\Delta^{14}\text{C}$ endmember. If the definition of the permafrost OC $\Delta^{14}\text{C}$ endmember is not straightforward, mass balance approaches used to study the relative contribution of different OC sources to Kongsfjord sedimentary OC similar to the approach of Kim et al. (2011) may lead to biased quantitative estimates. This implies that permafrost export (qualitatively and quantitatively) through time should be reconstructed from more than one single sedimentary record. Moreover, additional complications may arise if the permafrost OC $\Delta^{14}\text{C}$ endmember varies through time, for example, in response to changes in water mass distribution or circulation influencing hydrodynamic sorting processes or following erosion of different permafrost soil layers in response to changing temperature.

6. Conclusions

In this study, we use compound-specific ^{14}C analysis of alkanolic acids from soils, river, and fjord sediments from the Bayelva catchment and Kongsfjord. Our data demonstrate that bulk OC $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values from these samples are strongly biased by the contribution of coal particles. In contrast, alkanolic acid $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values reflect the carbon isotopic composition of vascular plant-derived permafrost OC and reveal that turnover in the Leirhaugen soil profile decreases with depth and is primarily determined by freeze-locking, that is, alkanolic acid ^{14}C ages are a function of limited microbial decomposition and radioactive decay. Depending on alkanolic acid chain length and soil depth, turnover happens on a multi-millennial scale as implied by ^{14}C ages ranging from $1,800 \pm 50$ years for $\text{C}_{16:0}$ alkanolic acid at 0–30 cm depth to $8,050 \pm 85$ years for $\text{C}_{26:0}$ alkanolic acid at 30–60 cm depth and are significantly longer than in other environments. Long-chain alkanolic acid $\Delta^{14}\text{C}$ values allow tracing the export of permafrost OC into the Bayelva River and Kongsfjord sediments. Their sedimentary isotope composition reveals that substantial amounts of deep active layer/permafrost OC are deposited off the Bayelva river mouth. Significant $\Delta^{14}\text{C}$ offsets between alkanolic acids in near-shore and offshore sediments reveals that sediments in the central fjord either receive input from other permafrost areas or are affected by sediment sorting (grain size/density effects) and accumulation rates.

Conflict of Interest

The authors declare no conflict of interests.

Acknowledgments

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Data Availability Statement

The data presented in this manuscript are archived in the PANGAEA data repository (Kusch et al., 2020).

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