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Sources and Accumulation of Sediment and Particulate Organic Carbon in a Sub-Arctic Fjord-Estuary: ²¹⁰Pb, ¹³⁷Cs, and δ¹³C records from Lake Melville, Labrador

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ABSTRACT

The sources and distribution of sediment and particulate organic carbon (OC) to Lake Melville, Labrador were characterized to better understand impacts from climate and hydrological changes to the system. Mass accumulation rates (MARs) across the Lake Melville System (LMS) were established from fifteen sediment cores collected in 2013 and 2014 by fitting $^{210}\text{Pb}_{\text{ex}}$ profiles to a two-layer advection diffusion model. MAR's, validated using ^{137}Cs , varied between 0.04 and $0.41 \text{ g cm}^{-2} \text{ y}^{-1}$, and overall decreased with increasing distance from the Churchill River, which drains into Goose Bay, a western extension of Lake Melville. The Churchill River is the greatest source of sediment to the system but surprisingly, MARs were greatest in western Lake Melville rather than Goose Bay, reflecting the contribution of fine material carried eastward in the Churchill River plume and inputs from nearby tributaries. A comparison of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ inventories to expected atmospheric fallout (1.5 and 23.6 dpm cm^{-2} , respectively) in sediment across LMS suggests particles are largely sourced from the watershed. In eastern Lake Melville, elevated $^{210}\text{Pb}_{\text{ex}}$ inventories and marine OC point to particle scavenging of dissolved ^{210}Pb from inflowing marine water. A transient tracer mixing model was used to determine the depth in each core where $>90\%$ of sediment was deposited before and after hydroelectric development at Churchill Falls (1970) and applied to downcore profiles of OC and $\delta^{13}\text{C}_{\text{org}}$. We observed a significant increase of terrestrial OC to Lake Melville post 1970, which we interpret as change from climate or hydrology of the Churchill River.

Introduction

Coastal environments frequently have high sedimentation rates due to inorganic and organic inputs from land. These regions, therefore, are globally important for the burial of organic carbon (OC) (Muller-Karger et al., 2005; Smith et al., 2015; Smith and Hollibaugh, 1993). During the last century Arctic and sub-Arctic rivers have generally shifted toward earlier freshets, and the annual amount of freshwater discharge to northern coasts has changed (Déry et al., 2011; Déry and Wood, 2005; Lammers et al., 2001; Peterson et al., 2002; Vörösmarty et al., 2000). In Siberia and Alaska, river inflow has been increasing whereas in western Canada inflow has generally decreased. These sorts of changes lead to poorly understood shifts in the delivery of inorganic and organic suspended matter carried by runoff (Syvitski et al., 2005). Over the same time interval, the supply of terrigenous material to Arctic coasts has been augmented directly or indirectly by rising sea levels and coastal erosion (e.g., Rachold et al., 2000; Nicholls and Cazenave, 2010), loss of sea ice cover (Jones et al., 2009), and thawing of permafrost (Lantuit and Pollard, 2008; Payette et al., 2004), all consequences of climate change.

For many northern coasts in Canada and elsewhere, fjords provide an interface between the river estuary and the open ocean. The high surrounding relief associated with fjords implies high sediment yields (Milliman and Syvitski, 1992), and the steep-sided fjord bathymetry, constrained by sills, leads to efficient sediment trapping in basins (Bentley et al., 2012; Syvitski, 1987). These basins, therefore, are important sites of long-term burial for terrigenous and marine organic matter.

Lake Melville, located in central Labrador, Canada (Figure 1), is much like a fjord but classified as a fjard. Lake Melville was formed by the submergence of a valley that had been produced by glaciation on land. This process of formation leads to three key differences from the

more commonly studied fjords: the adjacent land generally has lower topographic relief; the bathymetry tends to be shallower, broader, and irregular; and the drainage basin tends to be forested and have deeper soils. Lake Melville has the largest watershed (125,000 km²) in Labrador, accounting for 45% of the total surface water runoff from Labrador's land mass (Figure 1a), and a shallow sill at its eastern end that permits restricted exchange with the ocean. These characteristics imply that Lake Melville's basins provide effective locations to trap both terrestrial and marine particles.

In addition to the above-mentioned general changes associated with climate change, upstream hydrology was altered in the Lake Melville drainage basin by the Smallwood Reservoir located on the Upper Churchill River and the diversion of rivers with unknown consequences on Lake Melville (Figure 1b). Additional, the first phase of flooding of a newer hydroelectric development on the lower Churchill River at Muskrat Falls took place in October 2016 with more extensive flooding expected in 2017.

As part of a large interdisciplinary research and monitoring program to establish baseline conditions and to develop a scientific basis for monitoring Lake Melville in the context of climate change and hydro development (*Avativut, Kanuittailinnivut: Our Environment, Our Health*), we collected fifteen box cores from Lake Melville's basins. ²¹⁰Pb and ¹³⁷Cs, together with a sediment mixing model, were used to construct a geochronology for the past 100-150 years. Sediment profiles were also measured for total organic carbon (TOC) concentration and composition ($\delta^{13}\text{C}$). Our objectives were to: 1) Provide a baseline dataset for recent sedimentation rate within Lake Melville sediments; 2) Quantify the relative importance of OC from terrigenous and marine sources within recently accumulating sediments using $\delta^{13}\text{C}$ measurements and 3) Seek evidence of recent change in the quantity or composition of

accumulating sediment or OC. We discuss the sediment records in the context of sources of carbon, recent climate change and hydrological development in the Lake Melville drainage basin.

Study area

Lake Melville is a subarctic fjard estuary located within the Grenville Province of central Labrador, Canada (53.55°N and 59.58°W) (Roberts et al., 2006) (Figure 1). Around Lake Melville, flat lowlands dominate the landscape, apart from the Mealy Mountain plateaux, which border the southern shoreline of the fjard (Greene, 1974; Syvitski and Lee, 1997). The Kenamu River is one of the main rivers that drains the plateaux and discharges directly into western Lake Melville. Covering an area of about 2,100 km², Lake Melville is connected to Groswater Bay and the Labrador Sea by a long, narrow passage called Rigolet Narrows, hereafter referred to as the Narrows. The width of the Narrows (2.8 km), together with a sill (~35 m deep), restrict water exchange between Lake Melville and Groswater Bay. At the west end of Lake Melville, Goose Bay Narrows leads into Goose Bay (Figure 1c). Here we will refer to Goose Bay and Lake Melville collectively as the Lake Melville System (LMS).

Lake Melville features four sedimentary depressions where accumulation is likely to occur (Figure 1c). Epinette Basin (~160 m deep) lies nearest the outflows of the Kenamu River and is the first major basin east of Goose Bay; Sebaskachu Trough (~120 m deep) occupies a sedimentary bench in the northwest corner of Lake Melville; and Grand and Mulligan Troughs form the major deep basin (~250 m) for the LMS (Syvitski and Lee, 1997).

On an annual basis, the LMS receives on average ~94 km³ y⁻¹ freshwater, predominantly from four major rivers: Churchill, Goose, Kenamu, and Northwest (Bobbitt and Akenhead, 1982). The Churchill River, which has been altered in its upper regions through impoundment in

the Smallwood Reservoir (constructed between 1966 and 1974) and the diversion of the Nuaskaupi River (1971), the second largest river in Labrador that also discharges into Lake Melville via Grand Lake, alone supplies $\sim 70 \text{ km}^3 \text{ y}^{-1}$ of freshwater to Lake Melville. Annual discharge of the Churchill River has increased slightly since hydroelectric development and the seasonality shifted (increased flows in winter, decreased flows in spring/early summer).

The large input of freshwater to the western end of the LMS results in strong stratification and estuarine flow seen as a brackish surface layer (top 10-20 m) flowing seaward atop a saline bottom layer flowing landward from Groswater Bay (Cardoso and deYoung, 2002). During spring, summer and winter, a strong density gradient (halocline), 5-20 m deep, limits entrainment of saline bottom water to the surface layer where salinities are typically <10 psu, while bottom water salinities vary from about 20 to 28 psu (Cardoso and deYoung, 2002; Lu et al., 2013; Prinsenberget al., 2011). In October, surface mixing and cooling temperatures increase surface water density, weakening the halocline and facilitating mixing between the two water masses. This late fall-early winter mixing is reflected by the salinity of sea ice across Lake Melville (c.f., Prinsenberget al., 2011), which begins to form in late November. Complete freeze up occurs by mid to late December while breakup usually takes place in late April to May. Model simulations suggest that intensified winter river discharge from the Churchill River after 1971 has caused areas near the mouth of the Churchill and Northwest Rivers to experience higher ice velocities, more ice cracking, and more frequent ice-free periods in winter (Demirov and deYoung, 2016). Strong tidal currents sustain open water year round in the Narrows and Groswater Bay ($\sim 50 \text{ km}$ offshore) (Prinsenberget al., 2011).

138 **Methods**

139 *Sample collection*

140 Sediment cores were collected 10-17 June 2013 and 17-22 October 2014 using a box
141 corer deployed from *R/V Nuliajuk* and *M/V What's Happening*, respectively. Upon successful
142 collection of an undisturbed box core, a core tube was slowly pressed into the sediment, capped
143 and removed from the box corer. Sediment cores were extruded and sectioned into 1 cm
144 intervals, homogenized, and frozen (-20°C). River suspended particulate matter (SPM) was also
145 collected to determine $\delta^{13}\text{C}_{\text{org}}$ composition. Surface water from the mouths of the Churchill,
146 Goose, and Northwest Rivers were filtered through pre-combusted Whatman® GF/F filters
147 (nominal pore size, 0.7 μm).

148 *Radiochemical analysis*

149 Sediment samples were freeze dried, homogenized, and sealed in petri dishes for >21
150 days to allow for secular equilibrium. The gamma emissions of radioisotopes ^{210}Pb and ^{137}Cs
151 were counted at the Environmental Radiochemistry Laboratory (ERL) at the University of
152 Manitoba (Murray et al., 1987). Total ^{210}Pb and ^{137}Cs were counted for 24-48 hours at 46.5 and
153 661 keV, respectively. Activities of ^{226}Ra (used to estimate background ^{210}Pb) were estimated by
154 counting its granddaughter isotope, ^{214}Pb (352 keV). Detector efficiency and self-absorption
155 were corrected by counting reference material distributed by the International Atomic Energy
156 Agency (IAEA) within the same geometry (i.e. petri dish) (Cutshall et al., 1983). The
157 reproducibility (coefficient of variation), determined by counting the same sample four times,
158 were 5.7, 8.8, and 3.8%, for ^{210}Pb , ^{137}Cs , and ^{226}Ra , respectively. Excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) was
159 calculated as the difference between total ^{210}Pb and ^{226}Ra activities.

160 *Calculation of sediment and mass accumulation rates*

Sedimentation velocity (ω_s , cm y^{-1}), the rate at which sediment accumulates on the sea floor, was estimated by using a one dimensional two-layer advection diffusion model that accounts for both biomixing and compaction with depth (Kuzyk et al., 2013; Kuzyk et al., 2015; Lavelle et al., 1985):

$$[1] \quad \omega_s (\delta C / \delta z) - (\delta / \delta z) * K_b (\delta C / \delta z) = -\lambda C$$

where C is the $^{210}\text{Pb}_{\text{ex}}$ activity (dpm cm^{-3}), z is depth (positive downward in cm), K_b is the diffusion coefficient or mixing rate ($\text{cm}^2 \text{y}^{-1}$) due to bioturbation, and λ is the radioisotope decay constant for ^{210}Pb (0.03114y^{-1}). In this model, the sediment profile is treated as a two-layer system with a surface mixed layer (SML) influenced by bioturbation (K_{b1}) overlaying a deeper sediment layer with minimal mixing ($K_{b2} = 0.01 \text{cm}^2 \text{y}^{-1}$). The SML depths were assigned visually where $^{210}\text{Pb}_{\text{ex}}$ profiles transitioned from a homogenized profile near the surface to an observed exponential decrease with depth (Bentley et al., 2012; Kuzyk et al., 2009; Lavelle, 1985). For each core, K_{b1} and C_0 (surface activity, dpm cm^{-3}) were obtained by adjusting these values to obtain the best least-square fitting between the measured $^{210}\text{Pb}_{\text{ex}}$ profiles and the model output (Lavelle et al., 1986). Assuming constant rate of supply, the sedimentation velocities (Eq. 2) was calculated from the slope (m) of the linear regression of $\text{Ln } [^{210}\text{Pb}_{\text{ex}}]$ versus depth (cm) below the SML.

$$[2] \quad \omega_s = -\lambda / m$$

From sedimentation velocity, the mass accumulation rate (MAR , $\text{g cm}^{-2} \text{y}^{-1}$) was calculated as follows:

$$[3] \quad \text{MAR} = \rho_s (1 - \phi) * \omega_s$$

where ρ_s is the dry sediment density estimated at 2.65 g cm^3 (Berner, 1971) and $1 - \phi$ is the volume fraction of sediment particles where ϕ is the mean porosity below SML (Burdige, 2006). Porosity (ϕ) was calculated by the difference between the wet and dry weights of each sediment core slice and were salt corrected according to bottom water conditions at the time of sampling (cf., Lavelle, 1985).

MARs were validated by applying a numerical advection-diffusion model to corresponding ^{137}Cs (dpm g^{-1}) activities using parameters (ω_s , SML, K_{bl} , and C_0) derived from $^{210}\text{Pb}_{ex}$ profiles. On the basis of historical ^{137}Cs fallout and similar temporal variations of ^{137}Cs atmospheric fallout outlined by Monetti (1996) and Wright et al. (1999), an input function was developed and compared to measured values of ^{137}Cs (cf., Kuzyk et al., 2013). Sediment velocities were adjusted, within 95% confidence limits, to give the best fits for both $^{210}\text{Pb}_{ex}$ and ^{137}Cs .

$^{210}\text{Pb}_{ex}$ and ^{137}Cs inventories

Inventories (Eq. 4) of $^{210}\text{Pb}_{ex}$ and ^{137}Cs were calculated by summing the mass-depth activities down-core until activity was no longer detected (e.g., Kuzyk et al., 2009):

$$[4] \quad I = \sum C_i m_i$$

where C (dpm g^{-1}) is the $^{210}\text{Pb}_{ex}$ or ^{137}Cs activity at depth and m_i is the mass-depth (g cm^{-2}) increment (i) corresponding to the depth interval. Mass-depth was calculated as:

$$[5] \quad \text{Mass-Depth} = \rho_s * (1 - \phi) * \Delta_z$$

where Δ_z is the thickness of the sediment core interval (cm).

202 Inventories within 30% of estimated surface fluxes ($\text{dpm cm}^{-2} \text{y}^{-1}$, Eq 6) were used to check
203 sediment velocities and MARs.

204 [6] Surface Flux = $\omega_s * C_0$

205 Assuming that direct $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs atmospheric fallout onto the water surface would
206 be uniform (within expected uncertainty), the expected ^{210}Pb inventory from atmospheric
207 deposition was estimated as 23.6 dpm cm^{-2} . This value was calculated by dividing the expected
208 atmospheric flux of $0.732 \text{ dpm cm}^{-2} \text{y}^{-1}$, established by Appleby and Oldfield (1992) from direct
209 measurements of $^{210}\text{Pb}_{\text{ex}}$ in rainfall across eastern North America, by the decay rate of ^{210}Pb ,
210 0.03114 y^{-1} . The expected ^{137}Cs inventory ($\sim 1.5 \text{ dpm cm}^{-2}$ for 2013 and 2014) from direct
211 atmospheric fallout was calculated using a numerical model developed by Walling and He
212 (2000).

213 *Total organic carbon and $\delta^{13}\text{C}_{\text{org}}$ measurements*

214 Total organic carbon (OC) content and $\delta^{13}\text{C}_{\text{org}}$ were measured on surface sediment ($\sim 0\text{-}1$
215 cm) and down ten cores, at the University of Manitoba's Stable Isotope for Innovative Research
216 Laboratory (SIFIR). OC was derived from the difference between total carbon (TC) and
217 carbonate carbon (inorganic carbon, IC) in pre-weighed dried sediment. Replicate analysis of QC
218 standards B2150 (TC) and AR1034 (IC) indicated a propagated precision for OC of $\pm 0.1\%$.
219 Isotope composition of OC ($\delta^{13}\text{C}$ relative to VPDB) was measured on dried, decarbonated
220 samples using an elemental analyser isotope-ratio mass spectrometer (EA-IRMS) at SIFIR along
221 with an international standard, USGS Green River shale SGR-1b. Replicate samples of SGR-1b
222 ($n=21$) yielded a precision of $\pm 0.1\%$.

223 *Calculation of the percentage of terrestrial and marine organic carbon*

224 The percentage of terrigenous OC and, by difference, marine OC was calculated in
225 surface sediment and down select cores by applying a simple two-endmember mixing model
226 outlined in Stein and Macdonald (2004):

$$227 \quad [7] \quad \text{OC}_{\text{terr}}(\%) = (\delta^{13}\text{C}_{\text{sample}} - \delta^{13}\text{C}_{\text{mar}}) / (\delta^{13}\text{C}_{\text{terr}} - \delta^{13}\text{C}_{\text{mar}}) \times \text{TOC}(\%)$$

228 where $\delta^{13}\text{C}_{\text{sample}}$ is the measured $\delta^{13}\text{C}_{\text{org}}$ (‰) of the sediment sample and $\delta^{13}\text{C}_{\text{mar}}$ and $\delta^{13}\text{C}_{\text{terr}}$ are
229 the terrestrial ($-31.2 \pm 1.66\text{‰}$ ($\pm 1\text{SD}$, $n=6$)) and marine ($-21.4 \pm 0.32\text{‰}$ ($\pm 1\text{SD}$, $n=5$)) end-
230 members. The terrestrial end-member was established as the discharge weighted mean of $\delta^{13}\text{C}_{\text{org}}$
231 in SPM filtered from the Churchill, Northwest, and Goose Rivers. The marine end-member was
232 estimated from previously published surface sedimentary $\delta^{13}\text{C}$ value in Groswater Bay (Tan and
233 Vilks, 1987 and Muzuka and Hillaire-Marcel, 1999).

234 *Modelling of organic carbon*

235 The occurrence of a SML indicates mixing between newly deposited sediment and
236 sediment deposited at a previous time, making it inappropriate to assign specific ages to depths
237 in the sediment core (Johannessen and Macdonald, 2012; Kuzyk, 2015). Similar to Bailey et al.
238 (2013), we applied a transient tracer method outlined by Johannessen et al. (2005) to core
239 profiles and estimated the depth ranges in a given core that were deposited *before* and *after* the
240 hydro development in the upper Churchill River (i.e., pre- and post-1970). The sections of each
241 core representing $\geq 90\%$ of sediment deposited pre- and post-1970 were determined in
242 MATLAB® by adding a tracer in 1970 and allowing the model to run until date of core
243 collection (2013 and 2014). The modelled tracer concentration within each sediment slice was
244 then compared with the tracer input concentration.

Results

Porosity profiles

In most cores, the porosity profiles decreased smoothly downwards to depths just below the SML, followed by relatively constant porosity for the remainder of the core. In core 35, the porosity profile was noisier below 7 cm corresponding to coarser particles observed during sectioning.

^{210}Pb , ^{226}Ra , and ^{137}Cs radiochemistry

Total activities of ^{210}Pb in most cores decreased downwards, eventually merging with background levels of ^{210}Pb supported by ^{226}Ra decay (Figure 3). Supported ^{210}Pb in all cores was 1.5-3 dpm g⁻¹ and was reached at depths ranging from 8 to 20 cm. Profiles of total ^{210}Pb in cores 26 and I11, both collected in Goose Bay, did not reach background levels (Figure 3). While total ^{210}Pb in core 26 began rapidly declining below 8 cm, activity in core I11 was nearly constant throughout the length of the core, possibly reflecting erosion from the strong currents and ship traffic at this site. With the exception of the surface activity, total ^{210}Pb in core 35 was also somewhat unvarying, decreasing only slightly from 5.4 dpm g⁻¹ at 3 cm to 3.2 dpm g⁻¹ at 20 cm and thus not quite reaching background levels.

Except for core I11, the $^{210}\text{Pb}_{\text{ex}}$ profiles all exhibited two-layer behaviour indicative of a SML in which activities were fairly uniform and, beneath that, a layer in which $^{210}\text{Pb}_{\text{ex}}$ decreased exponentially with depth (Figure 4). The thickness of the SML ranged between 4-9 cm with a mean thickness of 5.8 ± 0.5 cm (mean $\pm$ SE, n=14).

The activity profiles for ^{137}Cs displayed a general shape that included no activity at the bottom of the core, peak activity between 4 and 14 cm and decreasing activity above that (Figure 4). Subsurface peaks in ^{137}Cs activity ranged between 0.7 and 1.3 dpm g⁻¹. Activities in cores

DB6 and DB3 reached levels below detection limit at the surface. In cores I10 and 35, ^{137}Cs was homogeneous throughout the core, penetrating deeper than would be expected from the $^{210}\text{Pb}_{\text{ex}}$ profiles. Due to the proximity of these cores to sources of freshwater (Goose and Kenamu Rivers), ^{137}Cs profiles could be displaying post-depositional re-mobilization through pore water (cf., Oughton et al., 1997).

Sedimentation velocity and mass accumulation rates

Sedimentation velocity and mass accumulation rate (MAR) exhibited greater than five-fold variability across LMS, ranging between 0.06-0.42 cm y^{-1} and 0.04-0.41 $\text{g cm}^{-2} \text{y}^{-1}$, respectively (Figure 5 and Table 1). Sedimentation velocities and MARs were highest in the western end of Lake Melville, in shallow waters (49 m) near the mouth of the Kenamu River (core 35, 0.42 cm y^{-1}) and in nearby deeper waters (161 m) in Epinette Basin (core 34, 0.25 cm y^{-1}), with corresponding MARs of 0.41 and 0.20 $\text{g cm}^{-2} \text{y}^{-1}$, respectively. In deep waters (217 m and 213 m) of eastern Lake Melville (Mulligan Trough), core DB5 had the lowest sedimentation velocity and MAR at 0.06 cm y^{-1} and 0.04 $\text{g cm}^{-2} \text{y}^{-1}$, respectively (Figure 5). The average sedimentation velocity and MAR in Grand Trough ($0.14 \pm 0.06 \text{ cm y}^{-1}$ and $0.09 \pm 0.03 \text{ g cm}^{-2} \text{y}^{-1}$ ($\pm \text{SE}$, $n=2$), cores 33, I13, 30), in central Lake Melville, are nearly double those found in Mulligan Trough ($0.07 \pm 0.01 \text{ cm y}^{-1}$ and $0.05 \pm 0.01 \text{ g cm}^{-2} \text{y}^{-1}$, ($\pm \text{SE}$, $n=2$), cores MT1 and DB3). In Goose Bay, average sedimentation velocities and MARs are higher than in Lake Melville, with averages of $0.16 \pm 0.04 \text{ cm y}^{-1}$ and $0.22 \pm 0.01 \text{ g cm}^{-2} \text{y}^{-1}$ ($\pm \text{SE}$, $n=2$) (cores I10 and 26), respectively. A higher sedimentation velocity and MAR was found near the mouth of the Churchill River ($\sim 0.20 \text{ cm y}^{-1}$ and $\sim 0.27 \text{ g cm}^{-2} \text{y}^{-1}$, core 26) compared to the mouth of the Goose River ($\sim 0.12 \text{ cm y}^{-1}$ and $\sim 0.16 \text{ g cm}^{-2} \text{y}^{-1}$, core I10). The $^{210}\text{Pb}_{\text{ex}}$ profile in core I11 could not be used to establish a sedimentation velocity and MAR, while the ^{137}Cs profiles in cores I10

and 26 could not be used to validate sedimentation velocities in these cores and reported values should be viewed with caution.

Inventories of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs

Inventories of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs in the LMS (Table 1) averaged 29.4 ± 2.7 ($\pm$ SE, $n=15$) and 4.9 ± 0.7 dpm cm^{-2} ($\pm$ SE, $n=14$), respectively. Generally, $^{210}\text{Pb}_{\text{ex}}$ inventories were lower in the shallow western regions of the LMS and higher in the deeper eastern end of the basin. For example, near the outflows of the Goose (core I10), Northwest (core I12) and, particularly, the Sebaskachu (core 28B) Rivers, where water depths are ≤ 50 m, $^{210}\text{Pb}_{\text{ex}}$ inventories were 25.5, 23.3, and 13.8 dpm cm^{-2} , respectively. In contrast, cores DB6 and BW1, collected from greater depths in the eastern end of the LMS, showed the greatest $^{210}\text{Pb}_{\text{ex}}$ inventories at 55.9 ± 2.1 and 40.7 ± 0.2 dpm cm^{-2} , respectively. The latter values are nearly double the expected inventory from direct atmospheric fallout onto the lake surface (23.6 dpm g^{-1}). Some exceptions to the east-west trend include core DB5, collected from a water depth of 217 m in the Mulligan Trough, which had one of the lowest $^{210}\text{Pb}_{\text{ex}}$ inventories (19.0 ± 2.0 dpm cm^{-2}), while a nearby core (MT1) had a $^{210}\text{Pb}_{\text{ex}}$ inventory of 31.0 ± 0.9 dpm cm^{-2} . ^{137}Cs inventories were similar in these two cores (2.3 ± 0.3 and 2.9 ± 0.1 , respectively), and similar to the average ^{137}Cs inventory (3.2 ± 1.7 dpm cm^{-2} ($\pm$ SE, $n = 2$)) found in the Grand Trough.

In all cores except I13 and 28B, ^{137}Cs inventories exceeded the expected inventory from direct atmospheric fallout (~ 1.5 dpm cm^{-2}). ^{137}Cs inventories were greatest in the western end of the LMS, near river mouths (i.e., 9.6 ± 0.2 dpm cm^{-2} in core 35 near the Kenamu River outflow). In the eastern end of the LMS, ^{137}Cs inventories were lower than average (i.e., 2.2 ± 0.3 and 4.7 ± 0.2 dpm cm^{-2} , cores BW1 and DB6, respectively).

313 *Distribution and profiles of organic carbon and $\delta^{13}C$*

314 Average TOC in surficial sediment across the LMS was $1.1 \pm 0.1\%$ ($\pm$ SE, $n=14$), with
315 most of the TOC values falling near 1% but a couple of cores having exceptionally low values
316 (29, I11) and core DB6 having values near 2% (Table 2). $\delta^{13}C_{org}$ values in surficial sediment
317 were generally low in the west and high in the east. The values ranged from a minimum value
318 of $-27.9 \pm 0.1\text{‰}$ in Goose Bay (core I10) to $-24.2 \pm 0.1\text{‰}$ in the eastern end of Lake Melville
319 (core DB6) and reached $-22.1 \pm 0.1\text{‰}$ in Groswater Bay (Table 2).

320 Profiles of TOC and $\delta^{13}C_{org}$ are compared in Figure 6a. In seven of the ten cores collected
321 across the LMS, $\delta^{13}C_{org}$ decreases up-core. In most cases, the up-core decrease is stepwise rather
322 than gradual and minimum values occur in shallow subsurface sediment rather than surface
323 sediment. In some cores (e.g., I13), the subsurface minimum is very pronounced, followed by a
324 return to higher values (similar to those deep in the core) in the surface sediment. The major
325 exception to this trend is core 26 from Goose Bay, in which $\delta^{13}C_{org}$ increases up core.

326 Because of biomixing, core depths cannot be translated into dates; instead, three zones or
327 layers are shown (with shading) in each core. These zones were defined using a transient tracer
328 mixing model as described above. The upper shaded zone reflects sediment deposited
329 predominantly ($\geq 90\%$) post-1970, the lower shaded zone reflects sediment deposited
330 predominantly pre-1970 ($\geq 90\%$), and the white (unshaded) zone reflects a mixture of pre- and
331 post-1970 sediment. The changes in $\delta^{13}C_{org}$ from the lower shaded zones (pre-1970) to the upper
332 shaded zones (post-1970) are presented in Table 3. For seven cores, there is a pre-1970 to post-
333 1970 decrease in $\delta^{13}C_{org}$ of 0.05-0.68‰ (Table 3). For three of these seven cores, there are three
334 or more sediment sections in both the pre- and post-1970 zones and statistical analyses indicate
335 that the pre- to post-1970 decrease is significant (ANOVA: core 33, $p=0.08$; core DB6, $p=0.05$).

Only core 26, collected from Goose Bay, exhibits a pronounced (and statistically significant) pre-1970 to post-1970 increase in $\delta^{13}\text{C}_{\text{org}}$ (0.32‰) (Table 3).

In Figure 6b, the percentage of OC_{terr} that we derived from the TOC and $\delta^{13}\text{C}_{\text{org}}$ data together is shown. In almost all cores across Lake Melville, OC_{terr} increases up-core, whereas the percentage of OC_{mar} remains relatively constant, suggesting this trend is not a result of diagenesis (Figure 6b). This trend is particularly evident in cores I12, 34, DB3, 33, and 30 but only cores 34, DB3, and 33 have three or more sediment layers in the pre- and post-1970. Nevertheless, the average OC_{terr} increases from pre- to post-1970 by 5% for cores 34, DB3, and 33.

Discussion

Inputs of riverine sediment inferred from mass accumulation rates

Spatial trends show that the rate of sediment accumulation throughout the LMS is significantly related to distance from the mouth of the Churchill River ($R^2=0.37$, $p<0.05$) (see Figure 8), while water depth plays a secondary role ($R^2=0.28$, $p=0.05$) (not shown). Two explanations as to why water depth plays a secondary role in particle distribution are: 1) the LMS's basin is irregular with multiple channels, shoals, and basins that complicate direct focusing of sediment from shallow to greater depths and, 2) the large extent of the Churchill River plume likely overshadows the role water depth has in this system. During periods of high river discharge, suspended sediments are transported eastward in the Churchill River plume, passing through Goose Bay Narrows and into Lake Melville (Figure 7). Coarse materials delivered by the river appear to remain near the river mouth, as implied by very low porosity values in sediments in this area and an increase in porosity with distance from the river (Figure 8a). Coarse debris tends to be deposited very close to shore in many river-dominated systems (Bianchi et al., 2007; de Haas et al., 2002; Gordon and Goni, 2004). As might be anticipated

from the eastward transport of fine-grained suspended sediment, MAR generally decrease with increasing distance from the Churchill River mouth (Figure 8b). The effect of distance from the predominant sediment source to the LMS can also be seen in other sediment properties. For example, ^{210}Pb inventory in sediment increases (Figure 8c), probably reflecting dilution by terrestrial sediment, ^{137}Cs inventory decreases (Figure 8d), and the $\delta^{13}\text{C}$ composition of surface OC increases (Figure 8e).

This simple picture is complicated by supply of sediment from smaller rivers, from coastal erosion, and the suitability for deposition at any particular coring site. For example, the highest MAR was near the mouth of the Kenamu River (core 35, $0.42 \text{ g cm}^{-2} \text{ y}^{-1}$). This value greatly exceeds the MAR observed in Goose Bay (core 26, $0.27 \text{ g cm}^{-2} \text{ y}^{-1}$), which is surprising because Goose Bay is nearer the outflow of the Churchill River. The greater MAR at the core 35 site probably reflects both direct sediment inputs from the Kenamu River (discharge $\sim 100 \text{ m}^3 \text{ sec}^{-1}$) (Anderson, 1985; Bobbitt and Akenhead, 1982) and the deposition of sediment transported in suspension eastward in the Churchill River plume. Current velocities in Goose Bay Narrows are reportedly as great as $\sim 5 \text{ m s}^{-1}$ (Syvitski, 1990). Goose Bay may, therefore be less favourable for sediment deposition than beyond the narrows where the Lake Melville basin widens and deepens. The low and vertically uniform radioisotope profiles in cores I10 and I11 (Figure 3) also point to mixing and/or resuspension associated with strong currents in Goose Bay. At the core 35 location, TOC in surficial sediment was three times greater than at core I10 in Goose Bay (Table 2). This spatial pattern suggests that fine, organic-rich particles largely bypass Goose Bay and are instead carried eastward through Goose Bay Narrows in the plume during freshet (Figure 7) and deposited on the east side where the basin widens and deepens. The large plume clearly also contributes fine grained material to Epinette Basin (core 34), the first sedimentary

depression east of Goose Bay, where the MAR is higher than anywhere else in Lake Melville at $0.20 \text{ g cm}^{-2} \text{ y}^{-1}$.

An indication of the influence of the nearby Kenamu River (2 km) at core 35 site includes the relatively low and variable porosity below the SML (Table 1) and particularly below 7 cm in the core (Figure 2). This low porosity (0.63), together with coarser material observed while sectioning core 35, not observed in other cores, suggest input of coarser sediment from the nearby Kenamu River rather than other sources like ice rafting. Syvitski (1990) suggested that the Kenamu River carries a larger fraction of (coarse) bedload material than suspended sediment to its delta relative to the Churchill River; a coarse component is consistent with the Kenamu River draining the steeper terrain of the Mealy Mountains (Anderson, 1985). Turbidity currents and mass wasting events are also common along the steep walls of Lake Melville's troughs (cf., Syvitski and Lee, 1997) and the Kenamu Delta is no exception (Syvitski, 1990). These turbidity currents would transport coarser particles episodically from the delta to the deeper basin.

Indications of riverine inputs, focusing, and boundary scavenging from ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ inventories

To evaluate the contribution of riverine sediment sources to the LMS, inventories of ^{137}Cs in the sediment cores were compared to the inventory attributable to direct atmospheric fallout ($\sim 1.5 \text{ dpm cm}^{-2}$). Higher ^{137}Cs inventories in sediments may be interpreted as ^{137}Cs that was previously deposited in the drainage basin and has entered the fjord on particulates delivered via river runoff (e.g., Smith et al., 2002). Unlike in marine water where an important component of ^{137}Cs from fallout exhibits conservative behaviour (Mackenzie et al., 1979), in terrestrial and freshwater environments, most of the ^{137}Cs binds almost irreversibly to clay particles by means of ion exchange (Jenne and Wahlberg, 1968; Smith and Ellis, 1982; Smith et al., 1987).

In the LMS, ubiquitous ^{137}Cs inventories $\geq 1.5 \text{ dpm cm}^{-2}$ imply a pervasive input of terrestrial ^{137}Cs . The spatial pattern in Figure 8d suggests that the Churchill River is an important source. Similar to MAR, an exceptionally high ^{137}Cs inventory (six times greater than expected from direct atmospheric fallout) was calculated for core 35 ($9.6 \pm 0.02 \text{ dpm cm}^{-2}$). Detectable activities of ^{137}Cs at the bottom of the core indicate that the actual inventory is probably larger than 9.6 dpm cm^{-2} . This large inventory integrates the combined contributions of fine material transported in suspension by the Churchill River plume, inputs from the Kenamu River, and direct deposition onto the water's surface. A relatively high ^{137}Cs inventory was also observed near the outflow of Northwest River (I12, $6.8 \pm 0.3 \text{ dpm cm}^{-2}$, Table 1).

The distribution of radioisotope inventories in estuarine and shelf environments is also affected by sediment focusing, which may occur through the lateral transport of fine grained material from shallow to deeper water (Crusius and Anderson, 1995; Hill et al., 2007). Following methodology developed by Kuzyk et al. (2013), we used the ^{210}Pb and ^{137}Cs inventories in tandem (Figure 9) to evaluate the relative importance of focusing (or lateral transport) and scavenging, driven by vertical particle flux (e.g., from primary production in surface waters). Focusing tends to enhance inventories of both ^{137}Cs and ^{210}Pb in sediments in a proportion that roughly reflects their ratio from atmospheric deposition. In contrast, scavenging tends to enhance inventories only for $^{210}\text{Pb}_{\text{ex}}$ in marine systems because $^{210}\text{Pb}_{\text{ex}}$ binds rapidly and almost irreversibly to suspended particles in saline water, which then settle to the seafloor (cf., Fang et al., 2013), whereas dissolved ^{137}Cs does not (Oguri et al., 2012).

Because of the strong enhancement of ^{137}Cs inventories in sediments influenced by river inputs (Figure 8d), here, the $^{210}\text{Pb}_{\text{ex}}/^{137}\text{Cs}$ inventory ratios in coastal sediments (<50 m water depth) vary from ~ 7.3 at sites (28B) remote from the river influence to ~ 3.6 at sites influenced by

the NW (I12), Goose (I10), Kenamu and Churchill (35) Rivers. A theoretical maximum $^{210}\text{Pb}_{\text{ex}}/^{137}\text{Cs}$ inventory ratio (~ 15.7) may be estimated from the expected atmospheric flux-derived inventories of ^{210}Pb (23.6 dpm cm^{-2}) and ^{137}Cs (1.5 dpm cm^{-2}). Thus, depending on the specific source sediments for focusing, this process may increase ^{137}Cs and ^{210}Pb inventories in sediments in ratios between 3.6 and 15.7 (see focusing vectors in Figure 9).

Figure 9 shows two broad groupings of cores with inventories that depart significantly from the expected atmospheric flux-derived inventories. In one group, which includes cores from more basinal sites in western Lake Melville (33, 34, DB3) and most of the coastal cores (35, I10, I12; water depth $< 50 \text{ m}$), the low ^{210}Pb and high ^{137}Cs inventories together indicate focusing from source sediments with strong river influence and/or direct influence of the Churchill (or another) River plume. These two processes, which both enrich ^{137}Cs inventories preferentially, cannot be distinguished without an additional tracer.

In the second group, which includes cores from eastern Lake Melville (MT1, I13, DB6) and the Backway (BW1), the high ^{210}Pb and low to moderate ^{137}Cs inventories together indicate scavenging and/or focusing from source sediments with minimal river influence. Across all the cores, $^{210}\text{Pb}_{\text{ex}}$ inventories generally increase eastward from the Churchill River ($R^2=0.27$, $p=0.06$, $n=14$; Figure 8c), a trend which may reflect scavenging of $^{210}\text{Pb}_{\text{ex}}$ from the in-flowing seawater from Groswater Bay (Radakovitch et al., 2003; Smoak et al., 1996). Alternatively, the higher $^{210}\text{Pb}_{\text{ex}}$ inventories in sediments accumulating at the eastern end of the LMS could also reflect higher primary production. It is well known that $^{210}\text{Pb}_{\text{ex}}$ is more effectively scavenged by particulate organic matter than fine lithogenic particles (i.e., clay) (cf., Moore and Dymondt, 1988; Oguri et al., 2012; Robbins and Edgington, 1975; Yang et al., 2015). In August 2012, Lu et al. (2013) recorded an upwelling event east of the sill in the Narrows. This renewal of deep,

cold water and its entry into the eastern end of Lake Melville, driven partly by estuarine entrainment, would provide an effective means to supply both dissolved ^{210}Pb from outside Lake Melville, and nutrients to support primary production within Lake Melville to scavenge it.

In the constricted eastern end of Lake Melville (core DB6), where $^{210}\text{Pb}_{\text{ex}}$ inventory is greatest (core DB6, $55.9 \pm 2.1 \text{ dpm cm}^{-2}$), the corresponding TOC (%) in surface sediment is greater than anywhere else in the LMS ($1.7 \pm 0.01\%$) (Table 2). In this area, the $\delta^{13}\text{C}$ values imply that ~70% of the TOC is OC_{mar} , which is more than double that of the OC_{mar} accumulating in Goose Bay cores. A positive association between percentage of OC_{mar} in sediment cores and $^{210}\text{Pb}_{\text{ex}}$ inventories ($R^2=0.43$, $p<0.05$, $n=10$) further supports the notion that scavenging of dissolved ^{210}Pb from inflowing seawater by vertical flux of marine organic matter (phytoplankton production) is an important process in the eastern end of Lake Melville.

Terrestrial and marine influence on sediment composition inferred from $\delta^{13}\text{C}_{\text{org}}$

The two-layer estuarine circulation of the LMS strongly influences the distribution and composition of particulates across the system as implied by the west to east trend of increasing $\delta^{13}\text{C}$ values for OC in surficial sediments along the axis of the LMS (Figure 8e). The implication of the isotope data is that the dominance of OC_{terr} sources delivered by river inflow at the western end of the LMS gives way to a dominance of OC_{mar} at the eastern end of the lake. This trend is partly because particles rich in OC_{terr} tend to settle out in the west and partly because primary productivity in the east is enhanced by entrainment and upwelling of deeper water over the sill in the Narrows (cf. Tan and Vilks (1987)).

In Goose Bay, $\delta^{13}\text{C}_{\text{org}}$ and OC (%) are lowest (i.e., -27.8‰ and 0.4%, respectively at site 26), reflecting the site's proximity to the major sources of terrestrial organic material and lithogenic material that would dilute the percentage of OC while lowering the $\delta^{13}\text{C}_{\text{org}}$ value. The

average $\delta^{13}\text{C}_{\text{org}}$ value in Goose Bay ($-27.8 \pm 0.03\text{‰}$ ($\pm\text{SE}$, $n=4$)) is 3.4‰ above the terrestrial endmember, suggesting some contribution of marine-derived OC in the western-most region of the Lake. In contrast, at the eastern end of Lake Melville near the Narrows and in Groswater Bay, $\delta^{13}\text{C}_{\text{org}}$ values in surface sediment ($-23.0 \pm 0.6\text{‰}$ ($\pm\text{SE}$, $n=3$)) are close to the marine endmember. These sediments contain a small amount of OC_{terr} , consistent with its distribution throughout surface waters of the fjord (Tan and Vilks (1987)).

Inferences of change in the sources of organic carbon to the Lake Melville System

The observed shifts within most cores towards lower $\delta^{13}\text{C}_{\text{org}}$ values and higher $\%\text{OC}_{\text{terr}}$ in more recently deposited sediments provide evidence that the sources and delivery of OC to the seafloor in the LMS have undergone change. The change implied by the up-core decrease in $\delta^{13}\text{C}_{\text{org}}$ values implies an increase over time in the relative contribution of terrestrial vs. marine OC sources, while the change implied by the up-core increase of $\%\text{OC}_{\text{terr}}$ is that the supply and burial of terrestrial OC has increased over time in these sediments. Although one core collected from Goose Bay showed an up-core increase in $\delta^{13}\text{C}_{\text{org}}$ (Figure 6a), this isotopic shift reflects the fact that both OC_{mar} and OC_{terr} increase upwards in that core, with the former slightly outpacing the latter (Figure 6b). Thus, even in this core, there is evidence of increased accumulation of terrestrial organic matter over time. Remineralization within the sediments can be ruled out as a significant contributor to the up-core trends in OC_{terr} because OC_{mar} , generally a more labile form of OC, is relatively invariant with some degree of burn down within the cores, while OC_{terr} shows independent trends.

There are several potential drivers of change in terrestrial OC delivery to sediments in the LMS, including altered hydrology or decreased sea ice duration and extent associated with climate change. Atmospheric warming has been recorded in the Labrador region since ~1998

(cf., Finnis and Bell, 2015; Way and Viau, 2014), but may well have been occurring for longer than that. Warming has been linked to changes in the type of OC delivered to coastal Arctic and sub-arctic environments over the last few decades through: i) alteration in river hydrology (e.g., Lammers et al., 2001; Roberts et al., 2012); ii) thawing of permafrost (e.g., Goni et al., 2005; Mann et al., 2015); and iii) longer open water season that favours greater fetch and storm surges (Lantuit et al., 2012), all of which could increase coastal and river bank erosion. In eastern Hudson Bay, an inferred offshore shift in sediment deposition during the past ~150 years determined from ^{210}Pb and ^{137}Cs sediment radiochemistry was attributed to a more energetic marine environment as a result of decreased sea-ice coverage (Hulse and Bentley, 2012). However, in contrast to eastern Hudson Bay, where the ice-free season has already lengthened by several weeks in recent decades, in the Lake Melville area of Labrador, recent sea ice timing, duration, and extent are still most strongly connected to climate variability, particularly the North Atlantic Oscillation (Finnis and Smith, 2016). Climate-related changes in ice cover therefore unlikely to explain the observed trends in OC_{terr} . Effects of a few unusually warm years in the last decade (particularly 2009/10 and 2010/11) probably would not be discernible in the sedimentary records examined here.

Hydroelectric development on the Upper Churchill River represents another possible driver of change in OC_{terr} accumulation in LMS sediments. To test the plausibility of this hypothesis, we examined whether the timing of changes in OC_{terr} accumulation in sediments in the LMS coincided with development of the Smallwood Reservoir and generating station at Churchill Falls. The finding of significant decreases in $\delta^{13}\text{C}_{\text{org}}$ and increases in OC_{terr} between the pre- and post-1970 time periods indicates that the timing of the change in the delivery of OC_{terr} to the LMS is not inconsistent with that of the hydrological developments.

Based solely on our data, we cannot determine the exact reason(s) for the augmentation of OC_{terr} accumulating in LMS sediments since 1970. But probable alternative to climate change, a plausible explanation is that the changes relate in some way to hydroelectric development on the Upper Churchill River. The Churchill River is the major source of sediment and OC_{terr} to Goose Bay and Lake Melville. Therefore, changes to the river could readily influence the delivery and type (i.e., soil organic material, fossil organic matter, fresh terrestrial plant material, and freshwater algae) of particulate OC_{terr} downstream (e.g., Kendall et al., 2001). Following the development of the Churchill Falls Generating Station and Smallwood Reservoir in the early 1970's, the mean annual discharge of the Churchill River increased by about $500 \text{ m}^3 \text{ y}^{-1}$ with the greatest increase ($\sim 1000 \text{ m}^3 \text{ y}^{-1}$) taking place December to March (Bobbitt and Akenhead, 1982; Syvitski, 1987). This increased discharge and change to seasonal runoff could have amplified OC_{terr} inputs to the system, at least for a period of time while the system readjusted (cf., Newbury and McCullough, 1984). Considering that changes in the Churchill River discharge triggered by the Upper Churchill hydroelectric development have influenced ice transport and thickness in parts of Lake Melville since 1971, increasing the occurrence of ice-free periods near the mouth of the Churchill River (Entcho and deYoung, personal communication), it is also possible that amplified OC_{terr} inputs were accompanied by enhanced dispersal within the LMS.

Due to the remoteness of Lake Melville, long term monitoring of the sediment and OC dynamics in this system does not exist. Therefore, it is not possible to conclude whether one or both of the above hypotheses is at play in the observed increase in OC_{terr} and decrease in $\delta^{13}C_{org}$ after 1970. With the expected completion of the Lower Churchill Hydroelectric Project in 2017, and ongoing climate change in the region (Finnis and Bell, 2015), long term monitoring of freshwater runoff, OC composition, and coastal erosion should be undertaken.

Conclusion

Most of the parameters measured in this study are associated to distance from the Churchill River, seen as gradients along the west to east axis of the LMS. For example, MARs are greatest in the western end of Lake Melville, immediately outside Goose Bay, where fine grained particles, transported in the plume of the Churchill River, are deposited. MARs decrease eastward along the LMS axis with some variation due to inputs from smaller rivers and local topographic lows (troughs, basins, and shallow narrow channels) that tend to trap re-suspended sediment.

Inventories of ^{137}Cs also decrease with distance from the mouth of the Churchill River. Inventories of ^{137}Cs either meet or exceed the estimated inventory from direct atmospheric deposition implying relatively large additional inputs of ^{137}Cs deposited within the drainage basin (i.e., through river runoff and coastal erosion) to the LMS. A comparison of $^{210}\text{Pb}_{\text{ex}}$ and corresponding ^{137}Cs inventories to the expected ratio in terrestrial soil reveals two groupings, those influenced by the Churchill and other river plumes and those influenced by particle scavenging of dissolved ^{210}Pb supplied by inflowing marine water.

Along the LMS axis, the trend in $\delta^{13}\text{C}_{\text{org}}$ in surficial sediments indicates that OC_{terr} dominates the western reaches but gives way to marine OC toward the eastern end of the LMS. This distribution reflects the large inflow of river water at the western end of the LMS and the resulting estuarine circulation, which entrains seawater entering at the Narrows. A statistically significant decrease in $\delta^{13}\text{C}_{\text{org}}$ value in sediments deposited predominantly (>90%) after 1970 implies that more OC_{terr} is presently entering the LMS than during historic periods. This increase could be a consequence of hydrological development, which was instituted during the 1970's, and climate change, which has likewise been occurring in this region.

In seeking reasons for the increase in OC_{terr} , it is likely that the strongest source of OC_{terr} would be involved. Specifically, changes in the flow and drainage area of the Churchill River because of hydroelectric development of Churchill Falls from 1967-1971 could have released OC_{terr} (cf., Houel et al., 2006), thus increasing the delivery of such OC_{terr} to the LSM at least for some period until the system readjusted. Global climate warming could also lead to an increase in freshwater runoff to the LMS, while at the same time, a shorter season of sea ice cover and greater storm events could increase coastal erosion and the delivery of OC_{terr} . Additionally, since winter discharge of the Churchill River has increased due to hydroelectric demand, the observed occurrence of ice-free periods near the mouth of the Churchill River has increased, which could amplify OC_{terr} inputs.

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Table 1: Sediment core properties, sedimentation rates, and parameters.

Core	Core Length (cm)	Water Depth (m)	Φ_{av}	SML (cm)	K_{b1} ($\text{cm}^2 \text{y}^{-1}$)	ω_s (cm y^{-1})	MAR ($\text{g cm}^{-2} \text{y}^{-1}$)	$\Sigma^{210}\text{Pb}_{ex}$ (dpm cm^{-2})	$\Sigma^{137}\text{Cs}$ (dpm cm^{-2})
I10 ^a	15	26	0.48	6	15	~0.12 (0.07-0.24)	~0.16 (0.10-0.33)	25.5±3.2	7.4±0.3
26 ^{a,b,c}	10	49	0.52	7	8	~0.20 (0.12-0.97)	~0.27 (0.14-1.18)	21.6±1.3	
I12 ^d	17	48	0.66	9	9	0.21 (0.02-0.43)	0.17 (0.02-0.41)	23.3±2.6	6.8±0.3
35 ^{b,d,e}	23	47	0.62	9	14	0.42 (0.15-0.82)	0.41 (0.18-0.79)	36.7±1.5	9.6±0.0
34 ^d	23	161	0.69	5	19	0.25 (0.18-0.27)	0.20 (0.14-0.22)	30.7±2.1	7.2±0.3
28B	14	50	0.69	4	3	0.09 (0.03-0.09)	0.07 (0.02-0.08)	13.8±1.2	1.9±0.4
DB3 ^d	17	126	0.70	5	6	0.18 (0.11-0.28)	0.12 (0.08-0.19)	25.3±5.2	5.4±0.6
MT1	18	149	0.73	7	3	0.07 (0.06-0.09)	0.05 (0.03-0.06)	31.0±0.9	2.9±0.1
DB5	14	217	0.74	4	3	0.06 (0.03-0.08)	0.04 (0.02-0.07)	19.0±2.0	2.3±0.3
33	20	175	0.76	7	9	0.19 (0.16-0.24)	0.11 (0.09-0.13)	34.1±1.9	4.9±0.2
I13 ^b	13	213	0.70	6	10	0.08 (0.02-0.08)	0.06 (0.02-0.07)	37.2±0.9	1.5±0.2
30	16	220	0.77	4	5	0.14 (0.08-0.40)	0.09 (0.04-0.25)	21.9±1.9	3.0±0.2
DB6	15	196	0.74	4	13	0.19 (0.12-0.41)	0.13 (0.09-0.28)	55.9±2.1	4.7±0.2
BW1	12	98	0.73	4	5	0.09 (0.05-0.12)	0.06 (0.04-0.08)	40.7±1.2	2.2±0.3

Note: SML= surface mixed layer; Φ_{av} = average porosity below SML; K_{b1} = upper layer mixing rate (mixing below K_{b1} , defined by K_{b2} (not shown), was 0.01 $\text{cm}^2 \text{y}^{-1}$ for all cores); ω_s = sedimentation velocity (95% confidence limits); MAR=mass accumulation rate (95% confidence limits); $\Sigma^{210}\text{Pb}_{ex}$ and $\Sigma^{137}\text{Cs}$ = sediment inventories ± propagated SD. ^aUnable to validate core due to incomplete or uniform ¹³⁷Cs profile; ^b²¹⁰Pb activities were determined by alpha method; ^cBackground levels were not reached within the length of the core. ^dMAR adjusted according to ¹³⁷Cs profile. ^e ω likely underestimated because ²¹⁰Pb activities at depth were variable, probably reflecting dilution from coarser grained particles.

Table 2: $\delta^{13}\text{C}_{\text{org}}$ (‰) and TOC (%) in surface sediment across the Lake Melville System and Groswater Bay.

Station	$\delta^{13}\text{C}_{\text{org}}$ (‰)	TOC (%)
TB1	-27.9	1.1
I10	-27.9	0.9
26	-27.9	0.4
I11	-27.8	0.5
I12	-26.8	0.8
35	-27.6	1.6
34	-27.5	0.9
28B	-26.4	1.1
DB3	-26.7	0.9
MT1	-26.3	1.3
33	-26.0	1.0
I13	-26.2	1.0
30	-25.0	1.1
DB6	-24.2	1.7
31	-22.8	1.4
32	-22.1	1.4

Table 3: Average $\delta^{13}\text{C}_{\text{org}}$ (‰) values in sediment deposited predominantly (>90%) pre- and post-1970 across the LMS and calculated change in $\delta^{13}\text{C}_{\text{org}}$ (‰) between these sedimentary zones.

Core	Average (n) $\delta^{13}\text{C}_{\text{org}}$ (‰)		Change ¹ (‰)
	Pre-1970	Post-1970	
I10	-27.93 (4)	-27.90 (1)	0.03
26	-28.22 (4)	-27.90 (1)	0.32
I12	-26.70 (5)	-26.75 (1)	-0.05
34	-26.90 (4)	-27.44 (2)	-0.54
MT1	-26.06 (7)	-26.25 (2)	-0.19
DB3	-26.30 (4)	-26.60 (3)	-0.30
33	-25.60 (12)	-25.91 (4)	-0.31 ²
I13	-26.33 (5)	-26.20 (1)	0.13
30	-24.76 (6)	-25.08 (2)	-0.32
DB6	-23.68 (4)	-24.36 (4)	-0.68 ²
Overall Average Change			-0.19

¹Positive change reflects increase in $\delta^{13}\text{C}_{\text{org}}$ from pre-1970 to post-1970, negative change reflects decrease in $\delta^{13}\text{C}_{\text{org}}$ from pre-1970 to post-1970; ²Significant at p=0.1 level

Maps showing the watershed of Labrador (a), location of Lake Melville relative to the Smallwood Reservoir (b), and Hamilton Inlet including Groswater Bay, Lake Melville and Goose Bay and major inflowing rivers (Churchill, Goose, Northwest, and Kenamu) (c).

123x155mm (150 x 150 DPI)

Porosity profiles of sediment cores. Observed coarse particles in slice 20 and 21 cm of core 35 are shown as open dots. These sections had unusually low ^{210}Pb activities and were omitted from modelling. The depth of the surface mixed layer in each core is shown by the horizontal dash line.

154x186mm (150 x 150 DPI)

Profiles of total ^{210}Pb (black dot) and ^{226}Ra (open dot) activities for cores collected in 2013 and 2014. Total ^{210}Pb activities in cores 26, I13, and MT1 were counted indirectly by alpha. The depth of the surface mixed layer in each core is shown by the horizontal dash line.

153x186mm (150 x 150 DPI)

<https://mc06.manuscriptcentral.com/cjes-pubs>

Sedimentation velocities (ω_s , cm y^{-1}) and mass accumulation rates (MAR, $\text{g cm}^{-2} \text{y}^{-1}$) across the Lake Melville System.

125x92mm (300 x 300 DPI)

Profiles of total organic carbon and $\delta^{13}\text{C}_{\text{org}}$ (a) and derived percentage of terrestrial and marine organic carbon (b) down sediment cores. Shading represents the section of the core with 90% of sediment deposited pre-1970 (lower shaded layer) and post-1970 (upper shaded layer). The non-shaded (middle) layers of the cores reflected a mixture of sediment deposited both pre- and post-1970.

180x108mm (300 x 300 DPI)

Plume of suspended sediment transported east from the mouth of the Churchill River during freshet. Image modified from NASA (NewfoundlandA2003141.1650.250m.jpg). Data Date: May 21, 2003; Visualization Date: May 29, 2003; Sensor(s): Aqua - MODIS

128x82mm (150 x 150 DPI)

Average porosity below SML (a), MAR(b), $^{210}\text{Pb}_{\text{ex}}$ inventories (c), ^{137}Cs inventories (d), and $\delta^{13}\text{C}$ and OC in surficial sediments (e) plotted against the distance to the Churchill River mouth (km).

88x210mm (150 x 150 DPI)

Inventories of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs in relation to the expected effects of three particle transport processes (scavenging, focusing, river plume influence) and the expected inventory composition from direct atmospheric deposition (X), are shown. Cores with $^{210}\text{Pb}/^{137}\text{Cs}$ ratios that fall in the right grouping (i.e., high ^{137}Cs inventories) have been influenced by focusing from sediments strongly influenced by river discharge and/or direct influence of the Churchill River plume. Cores in the left grouping have been influenced by focusing from source sediments with minimal river influence and/or scavenging of $^{210}\text{Pb}_{\text{ex}}$ from in-flowing seawater from Groswater Bay.

162x143mm (150 x 150 DPI)

Supporting Information

Sources and Accumulation of Sediment and Particulate Organic Carbon in a Sub-Arctic
Fjord-Estuary: ²¹⁰Pb, ¹³⁷Cs, and δ¹³C records from Lake Melville, Labrador

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Methods

Sample collection

Sediment core sites were selected by reference to Canadian Hydrographical Service nautical charts (4724, 4728, 5140, and 5143), multi-beam bathymetric data collected and processed by the Ocean Mapping Group at the University of New Brunswick in 2012 and 2013, and previously published literature. Areas of interest for sediment core collection included the basins (>100 m), where sediment deposition was likely to be important, and near river mouths where the greatest input of particulates was expected to occur. On retrieval, sediment cores were visually inspected: if the sediment appeared disturbed, it was discarded and the box core was re-deployed. Upon successful collection of an undisturbed box core, excess overlying water was siphoned off with care to ensure minimal disturbance to the sediment. A 10 cm diameter plastic tube was used to subsample the box core by pressing it slowly into the sediment ≥ 5 cm from the box edge and subsequently capping both ends. Sediment cores were extruded and sectioned into 1 cm intervals, placed in Whirl-Pak® bags, homogenised by hand, and frozen (-20°C).

Suspended particulate matter (SPM) was collected near the mouths of the Churchill, Goose, and Northwest Rivers in June 2013 and October 2014 to determine $\delta^{13}\text{C}_{\text{org}}$ composition. From the shore, surface water (top ~0.25-0.5 m) was pumped for 0.5 to 1 hour through a Teflon-lined hose connected to a stainless steel filter holder, to collect particulates on pre-combusted Whatman® GF/F filters (nominal pore size, 0.7 μm).

Radiochemical analysis

To determine sedimentation rates, the activities of the radioisotopes ^{210}Pb and ^{137}Cs were measured in sediment core sections at the Environmental Radiochemistry Laboratory (ERL) at the University of Manitoba. Sediment samples were prepared by thawing, re-homogenising samples by hand, sub-sampling, refreezing, and freeze drying sediment sections at -55°C for approximately 4 days. To estimate ^{210}Pb supported by ^{226}Ra decay in the sediments, freeze dried samples were ground using a pestle and mortar and sealed in Petri dishes for ≥ 20 days to prevent the escape of ^{222}Rn and allow secular equilibrium with ^{214}Pb .

Weighed and sealed samples were measured for total ^{210}Pb and ^{137}Cs by counting for 24-48 hours the gamma emissions at 46.5 and 661 keV, respectively, on a CANBERRA® Broad Energy Germanium (BEGe) P-type detector (see Zaborska et al., 2007). Activities of ^{226}Ra were estimated by counting its granddaughter isotope, ^{214}Pb , at 352 keV. Detector efficiency and self-absorption were corrected by counting reference material distributed by the International Atomic Energy Agency (IAEA) within the same geometry (i.e. petri dish) (Cutshall et al., 1984). The reproducibility (coefficient of variation), determined by counting the same sample four times, were 5.7, 8.8, and 3.8%, for ^{210}Pb , ^{137}Cs , and ^{226}Ra , respectively. To assess the appropriateness of counting total ^{210}Pb directly by gamma emission in sediment samples from the LMS, four sediment cores (26, I13, DB3, MT1) were subjected to the alpha technique (Flynn, 1968). The reproducibility error, determined by counting the same sample four times, was less than 1%. Excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) was calculated as the difference between total ^{210}Pb and ^{226}Ra activities.

Calculation of sediment and mass accumulation rates

Sedimentation velocity (ω_s , cm yr^{-1}), which is the rate at which sediment settles to the sea floor, was estimated by using a one dimensional two-layer advection diffusion model that accounts for both bioturbation and compaction with depth (Kuzyk et al., 2013; Kuzyk et al., 2015; Lavelle et al., 1985):

$$[1] \quad \omega_s (\delta C / \delta z) - (\delta / \delta z) * K_b (\delta C / \delta z) = -\lambda C$$

where C is the $^{210}\text{Pb}_{\text{ex}}$ activity (dpm cm^{-3}), z is depth (positive downward in cm), K_b is the diffusion coefficient or mixing rate ($\text{cm}^2 \text{yr}^{-1}$) due to bioturbation, and λ is the radioisotope decay constant (0.03114yr^{-1}). In this model, the sediment profile is treated as a two-layer system with a surface mixed layer (SML) influenced by bioturbation (K_{b1}) overlaying a deeper sediment layer with minimal mixing ($K_{b2} = 0.01 \text{cm}^2 \text{yr}^{-1}$). The SML depths were assigned visually by choosing inflection points down the core profiles where $^{210}\text{Pb}_{\text{ex}}$ activity transitioned from a homogenized profile near the surface to an observed exponentially decrease with depth (Bentley et al., 2012; Kuzyk et al., 2009; Lavelle, 1985). For each core, K_{b1} and C_0 (surface activity, dpm cm^{-3}) were obtained by adjusting these values until the best least-square fitting between the measured $^{210}\text{Pb}_{\text{ex}}$ profiles and the model output. Assuming constant rate of supply, the sedimentation velocities (Eq. 2) were (Johannessen and Macdonald, 2012) calculated from the slope (m) of the linear regression of $\text{Ln } ^{210}\text{Pb}_{\text{ex}}$ versus depth (cm) below the SML.

$$[2] \quad \omega_s = -\lambda / m$$

From sedimentation velocity, the mass accumulation rate (MAR, $\text{g cm}^{-2} \text{yr}^{-1}$) was calculated as follows:

$$[3] \quad \text{MAR} = \rho_s (1 - \Phi) * \omega_s$$

where ρ_s is the dry sediment density estimated at 2.65g cm^{-3} , the density of quartz and commonly applied to marine sediment (Berner, 1971) and $1 - \Phi$ is the volume fraction of sediment particles where ϕ is the mean porosity below SML (Burdige, 2006).

MARs were validated by applying a numerical advection-diffusion model to corresponding ^{137}Cs (dpm g^{-1}) activities using parameters (ω_s , SML, K_{b1} , and C_0) derived from $^{210}\text{Pb}_{\text{ex}}$ profiles. On the basis of

historical ^{137}Cs fallout and similar temporal variations of ^{137}Cs atmospheric fallout outlined by Monetti (1996) and Wright et al. (1999), an input function was developed and compared to measured values of ^{137}Cs (cf., Kuzyk et al., 2013). The model was run in MATLAB® and accounted for radio-decay of ^{137}Cs (0.0231 yr^{-1}), plausible delayed inputs from drainage basins (Walling and Qingping, 1992), post depositional remobilization such as bioturbation and molecular diffusion (Robbins and Edgington, 1975) and/or sediment focusing and resuspension of deposited sediment (Crusius and Anderson, 1995). Sediment velocities were adjusted, within 95% confidence limits, to give the best fits for both $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs .

$^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs inventories

Inventories (Eq. 4) of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs were calculated by summing the mass-depth activities down-core until activity was no longer detected (e.g., Kuzyk et al., 2009):

$$[4] \quad I = \sum C_i m_i$$

where C (dpm g^{-1}) is the $^{210}\text{Pb}_{\text{ex}}$ or ^{137}Cs activity at depth and m_i is the mass-depth (g cm^{-2}) increment (i) corresponding to the depth interval. Mass-depth was calculated as:

$$[5] \quad \text{Mass-Depth} = \rho_s * (1 - \Phi) * \Delta_z$$

where Δ_z is the thickness of the sediment core interval (cm).

Inventories within 30% of estimated surface fluxes ($\text{dpm cm}^{-2} \text{ yr}^{-1}$, Eq 6) were used to check sediment velocities and MARs.

$$[6] \quad \text{Surface Flux} = \omega_s * C_0$$

we assumed that direct $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs atmospheric fallout onto the water surface would be uniform (within expected uncertainty) across the LMS. The expected ^{210}Pb inventory accumulated from atmospheric deposition was estimated as 23.6 dpm cm^{-2} . This value was calculated by dividing the expected atmospheric flux of $0.732 \text{ dpm cm}^{-2} \text{ yr}^{-1}$, established by Appleby and Oldfield (1992) from direct

measurements of $^{210}\text{Pb}_{\text{ex}}$ in rainfall across eastern North America, by the decay rate of ^{210}Pb , 0.03114 y^{-1} .

The expected ^{137}Cs inventory ($\sim 1.5 \text{ dpm cm}^{-2}$ for 2013 and 2014) from direct atmospheric fallout was calculated using a numerical model developed by Walling and He (2000).

Total Organic carbon and $\delta^{13}\text{C}_{\text{org}}$ measurements

Total organic carbon (TOC) content and $\delta^{13}\text{C}_{\text{org}}$ were measured on surface sediment ($\sim 0\text{--}1 \text{ cm}$) and down ten cores, at the University of Manitoba's Stable Isotope for Innovative Research Laboratory (SIFIR). TOC was derived from the difference between total carbon (TC) and carbonate carbon (inorganic carbon, IC). Both TC and IC were measured from pre-weighed dried sediment subsamples. TC was measured by combusting the subsample at $\sim 1400^\circ\text{C}$ and analysing using an elemental analyser. IC was measured by removing inorganic carbon (decarbonation) from the subsample with 20% HCl and immediately analysing the gas released from the sample using a CO_2 coulometer. Replicate analysis of QC standards B2150 (TC) and AR1034 (IC) indicated a propagated precision for OC of $\pm 0.1\%$.

Organic carbon isotope composition ($\delta^{13}\text{C}$ relative to VPDB) was measured on dried, decarbonated samples using an elemental analyser isotope-ratio mass spectrometer (EA-IRMS) at SIFIR. To ensure quality of sample (decarbonation) and analysis performance, an international standard, USGS Green River shale SGR-1b, was treated and analysed along with samples. Replicate samples of SGR-1b ($n=21$) yielded a precision of $\pm 0.1\%$.

Calculation of the percentage of terrestrial and marine organic carbon

The percentage of terrigenous OC and, by difference, marine OC were calculated for each core and surface sediment by applying a simple two-endmember mixing model outlined in (Stein and Macdonald, 2004):

$$[\text{7}] \quad \text{OC}_{\text{terr}}(\%) = (\delta^{13}\text{C}_{\text{sample}} - \delta^{13}\text{C}_{\text{mar}}) / (\delta^{13}\text{C}_{\text{terr}} - \delta^{13}\text{C}_{\text{mar}}) \times \text{TOC}(\%)$$

where $\delta^{13}\text{C}_{\text{sample}}$ is the measured $\delta^{13}\text{C}_{\text{org}}$ (‰) of the sediment sample and $\delta^{13}\text{C}_{\text{mar}}$ and $\delta^{13}\text{C}_{\text{terr}}$ are the terrestrial ($-31.2 \pm 1.66\text{‰}$ ($\pm 1\text{SD}$, $n=6$)) and marine ($-21.4 \pm 0.32\text{‰}$ ($\pm 1\text{SD}$, $n=5$)) end-members. The terrestrial end-member was established as the discharge weighted mean of $\delta^{13}\text{C}_{\text{org}}$ in SPM filtered from

the Churchill, Northwest, and Goose Rivers. Because of mixing between fresh and marine water in the Narrows, the marine end-member was difficult to establish. We used the mean and standard deviation of previously published data of $\delta^{13}\text{C}$ signatures from surficial sediment in Groswater Bay reported by Tan and Vilks (1987). Muzuka and Hillaire-Marcel (1999) also reported similar signatures for surficial sediment east of Groswater Bay.

Modelling of organic carbon

The occurrence of a SML indicates mixing between newly deposited sediment and sediment deposited at a previous time, making it inappropriate to assign specific ages to depths in the sediment core (Johannessen and Macdonald, 2012; Kuzyk, 2015). Similar to Bailey et al. (2013), we applied a transient tracer method outlined by Johannessen et al. (2005) to core profiles and estimated the depth ranges in a given core that were deposited *before* and *after* the hydro developments in the upper Churchill River (i.e., pre- and post-1970). The sections of each core representing $\geq 90\%$ of sediment deposited pre- and post-1970 were determined in MATLAB® by adding a tracer in 1970 and allowing the model to run until date of core collection (2013 and 2014). The modelled tracer concentration within each sediment slice was then compared with the tracer input concentration.

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