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Memo

To: Jackie Wells, EA Commitment/Environmental Effects Monitoring Programs Lead

From: James McCarthy, Senior Biologist/Ecosystem Insight Group Lead

cc: Matthew Gosse, Environmental Biologist

Date: September 1, 2020

Re. Sediment Sampling Results, Goose Bay and Lake Melville

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1. Introduction

Sediment samples collected prior to 2019 from Goose Bay and Lake Melville have been ineffective to generate samples with appropriate sediment types to detect methylmercury. As part of their review process of the Environmental Effects Monitoring (EEM) Program, the Independent Expert Committee (IEC) appointed by the provincial government provided recommendations related to the sampling of sediment from Goose Bay and Lake Melville. It specifically recommended replacing existing sediment sampling using an Eckman sampler, with collection of intact dated sediment cores using a gravity corer:

> *"Mercury in sediment is primarily analyzed to provide information on depositional processes of mercury and the pool of mercury potentially available for transfer into the water column of the reservoir and export further downstream. However, the current method of sediment collection (Eckman sampler) disturbs sediment stratification and, thus, information on the timeline of mercury deposition. Thus, dated lake sediment cores are widely used to examine changes in deposition of mercury and other contaminants over time."*
>
> *"We recommend sampling sediment cores from all sampling locations in the reservoir, Goose Bay and Lake Melville (sediment cores from rivers can be problematic due to the rapid movement of sediment) once in summer 2018. These cores would be sliced at 0.5 cm intervals and slices would then be dated using lead-210 methods and analyzed for concentrations of THg. We recommend that these sites be re-cored every ~5 years to examine changes in sedimentation rates and mercury deposition over time. We recommend that only the top 5 cm of each core be analyzed in samples collected every 5 years. We can make a more refined recommendation (sampling resolution, sample frequency) upon consultation with Zou Zou Kuzyk at the University of Winnipeg who cored similar sites and has determined sedimentation rates."*

While this recommendation provided general advice, no additional details or follow up was provided. Therefore, a detailed methodology was developed for sediment collection to acquire the methylmercury and aging (Pb-210) data as requested and submitted prior to sample collection in May 2019. In order to ensure the most reliable design for collection of depth-stratified samples, the general and site-specific challenges related to this program were reviewed and addressed within the methodology. Provided below is a summary of the method and the results of the sediment sample analysis.

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2. Methodology

Sediment samples can reveal chemical and metal contaminations in a time-integrated sample (Hakanson and Jansson 1983) provided they are collected in such a way as to preserve their structural integrity. They can also be used to determine sedimentation rates provided sample integrity is maintained (Robbins and Edgington 1975). While sediment samples can be relatively simple to collect to document rates of sedimentation and methylmercury concentrations (Håkanson and Jansson 2002); the nature of the lower Churchill River and the Goose Bay/Lake Melville estuary presents challenges with respect to appropriate, representative samples (and sites) False conclusions are an obvious risk if such studies are based on biased field samples (Blomqvist 1991).

2.1 Sample Collection

Gravity core samplers are typically used to sample vertical columns of sediment as they can maintain a more representative vertical profile of the sediment stratigraphy, create less disturbance by shock waves and can collect more highly consolidated deposits (Ohio EPA 2001, U.S. EPA 2014). They are particularly useful in pollutant monitoring because turbulence created by descent through the water is minimal (Håkanson and Jansson 2002), thus the fines at the sediment-water interface are only minimally disturbed; the sample is withdrawn intact, permitting the removal of only those layers of interest; core liners of glass or Teflon reduce possible sample interferences; and the samples are easily delivered to the lab for analysis (Region 4 US EPA Science and Ecosystem Support Division, Athens, Georgia. Sediment Sampling Operating Procedure 2014).

For this program, an 8 cm diameter HTH Corer with Teflon liners was used to collect sediment samples.

2.2 Sample Locations

Sample location is very important for obtaining representative samples with minimal disturbance, redistribution, and biological mixing. The sedimentological record in dynamic estuarine environments is often distorted or obscured by sediment reworking as a result of biological activity or by depositional and erosional processes associated with phenomena such as wave action or storm surges (Smith and Walton 1980). Additionally, sites in the deepest water may have faster rates of sediment accumulation because of resuspension of shallow-water sediment and its deposition in deep water (focusing) (Wright 1991).

Samples were collected from the deepest portions of Goose Bay and Lake Melville using the general bathymetry outlined in Figure 1. As shown, there are several locations where deeper troughs and shelves are identifiable, therefore four separate locations were included. Table 1 provides the coordinates of each sample location.

Samples were collected in May 2019 using the existing spring stable ice cover to assist in anchoring and maintaining vertical position during coring (Blomqvist 1991, Wright 1991). Using ice cover to maintain vertical position also allowed very controlled descent and retrieval of the corer. A mobile sonar was also deployed to determine distance to the bottom. The tether was marked at 20 m intervals so that during the last 20 m before the sediment interface, the unit could be slowly lowered into the substrate.

Each core was retrieved and the core liners containing the sediment sample removed and processed (sectioned) at the sample location prior to demobilizing to avoid resuspension of material and/or core mixing. Each sample was initially held in a vertical position and placed in a stand where it was inspected for integrity. Selected cores were sectioned, using a manual extruder, after the floc was allowed to resettle for up to an hour. The entire process was completed during the spring at which time the air and water temperatures were similar as possible so as to prevent sediment expansion due to gas formation. Any suspended floc remaining was removed by a syringe and kept for analysis, while the more consolidated sediments, intervals of well-defined thickness were isolated by means of a centimetre-scribed collar, placed over the end of the core liner, into which an appropriate amount of material was extruded.

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During the sampling period, all core sections for lab analysis were temporarily stored in coolers without ice (Wright 1991). Core sections were shipped immediately after the field program to the Flett Research lab for analysis. Photos of each core prior to extrusion were collected to document layering, colour, and condition (e.g., disturbance of surface layer.

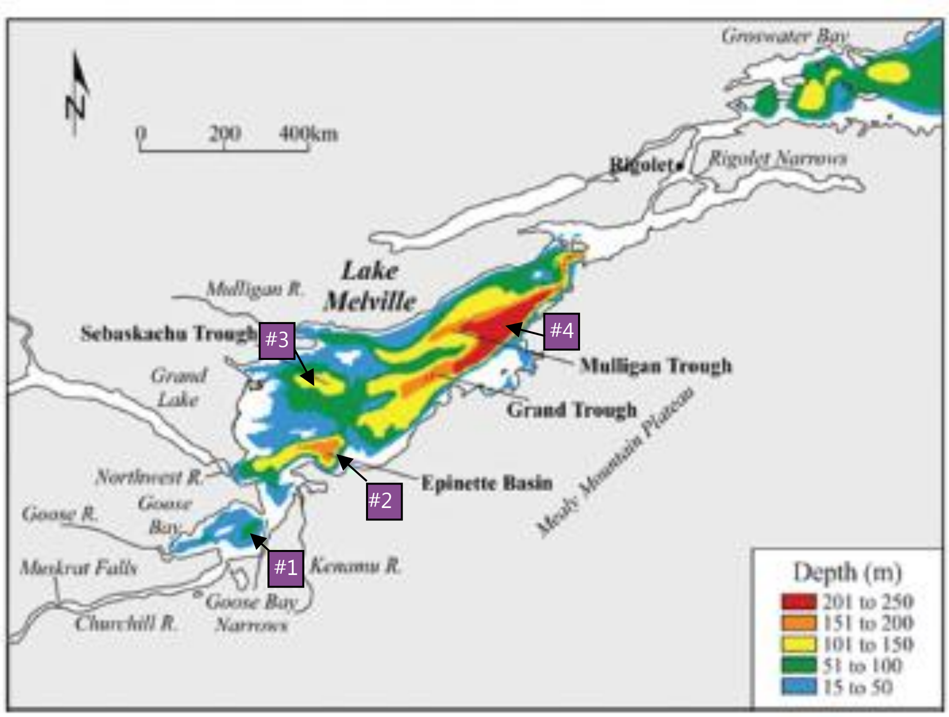

Figure 1: Bathymetric contours, Lake Melville (Kamula et al. 2017). Proposed sample locations are indicated with numbers 1-4.

Table 1: Sediment sample locations, Goose Bay and Lake Melville, May 2019.

Sample ID	Approximate Water Depth (m)	UTM Zone	Northing	Easting
GB03	37	20	5922806	691424
LM02	130	21	5935543	316726
LM03	95	21	5953583	312205
LM04	210	21	5959398	353967

Flett Research Labs indicate that the resolution of core sections depends upon the sediment accumulation rate at the sampling site. Typically, cores are sectioned at 1-cm thickness in the upper 20 cm, 2-cm thickness between 20 and 40 cm and 5-cm thickness below 40 cm depth. If the sediment accumulation rate is very low (e.g. <0.05 cm/yr), then a higher resolution would typically be desired (i.e. 0.5 cm / section). If the sediment accumulation rate is very high (e.g., in a dam or river inlet), the core can be sectioned at 2-cm or 5-cm thickness or even thicker. If these samples are to be analyzed for other analytes (e.g. metals or contaminants), then the resolution may need to be defined by the study. Smith and Walton (1980) showed that cores can be homogenously mixed within upper 4 – 5 cm due to bioturbation caused by benthic invertebrates and fish disturbing the sediment during activities. The top 5 cm of sediment cores previously collected by other researchers in Lake Melville have also shown bioturbation (M. Kamula pers comm.). Therefore, the recommended sampling and sectioning of only the upper 5cm by the IEAC will not likely provide information on settling rates or methylmercury accumulation within the sediment material.

Kamula et al. (2017) completed coring within Goose Bay and Lake Melville in areas similar to those proposed for this program. The typical core depths obtained were 10-23 cm in these areas with estimated sedimentation velocities of 0.14-0.25 cm/yr.

Based on the above previous observations and lab recommendation, each core was sectioned every centimetre throughout the entire core length collected to capture the available existing sediment dates and provide a description of the existing sediment mercury distribution. Each section sample was well homogenized and collected in clean, waterproof, heavy walled polypropylene jars. Jar rims were kept clean when closing to avoid particles on the sealing surfaces. Water loss from each section is to be avoided as this will affect the bulk density which in turn will affect the Pb-210 dating calculations. Samples did not require special storage conditions for radioisotope analyses; holding times are unlimited. However, Total Mercury analysis required shipment as soon as possible for analysis.

Samples were provided to the lab as wet samples so that dry bulk density of the sediment (dry wt./wet volume) could be calculated before drying and analyzed for Pb-210.

2.3 Sample Analysis

Total Mercury Concentration

Determination of Total Mercury (THg) in each sediment core section sample was completed using EPA Method 7473 for DMA-80 Total Mercury Analyser as performed by Flett Research Ltd. After removing a sub-sample of sediment for bulk density determination, samples were freeze dried and ground by hand prior to mercury analysis. The determined recordable detection limit (RDL) of analysis was 6 ng/g. The method detection limit (MDL) was determined to be 2 ng/g based on seven replicates of analytical blanks (98% confidence). This limit assumed 10 mg dry samples. Lower detection limits are possible if greater sample weights are used. The estimated uncertainty for this method is ±17% (95% confidence) at total mercury concentrations between 4 and 3,000 ng/g.

Date Estimation

Obtaining good cores and making the right radioisotopic measurements are very important in generating the best possible estimates of sediment age within a core (Flett Research). The Pb-210 method is used to determine the accumulation rate of sediments in lakes, oceans and other water bodies. In a typical application, the accumulation rate over a period of 100 - 200 years can be obtained. From the accumulation rate, the age of sediment from a particular depth in the sediment column can be estimated.

Sedimentation rates were completed using radioactive isotopes of Pb-210 and Cs-137 on the same core samples. The spatial distributions of both Cs-137 and Pb-210 are principally via attachment to setting particles (Robbins and Edgington 1975, Ritchie and McHenry 1990) and are often used in combination to provide information on soil and sediment redistribution over the last 100-year time period. Since the 1970s, Pb-210 measurements have been used extensively for dating sediment deposits in a range of sedimentary environments, including, lakes, reservoirs, flood plains, wetlands, estuaries, and coastal marine environments, thereby permitting sedimentation rates of the last 10-150 years to be determined (Robbins and Edgington 1975, Kristensen et al. 2012, Mabit et al. 2014). When applying the Pb-210 technique, it is assumed that lake and ocean sediments are receiving a constant input of Pb-210 from the atmosphere. Cs-137 is radioactive fallout deposited across landscapes from atmospheric nuclear tests that is strongly absorbed on soil particles, limiting its movement by chemical and biological processes (Ritchie and McHenry 1990).

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As a result, most Cs-137 movement in the environment is by physical processes and hence is a unique tracer for studying erosion and sedimentation.

Since all cores were less than 15-20 cm in length, the entire set of core sections were analyzed for Pb-210 for each sample. Several (two to three) Ra-226 measurements are often required for each core, in order to positively determine the Pb-210 background level(s). After all Pb-210 and Ra-226 analyses were completed, Cs-137 analyses were completed for each core, in order to validate the Pb-210 model(s).

Pb-210 dating was completed using N20110 Determination of Lead-210 in Sediment, Soil, and Peat by Alpha Spectrometry. The MDL for 0.25-0.5 g (dry wt.) sample was between 0.005-0.1 DPM Po-210/g dry sample at a 95% confidence level of 60,000 second counting time and is based on greater than 20 method blanks. This can vary slightly and depends upon the amount of sample, detector, and recovery efficiency of each sample. The estimated uncertainty for samples analyzed by this method (acid extraction) has been determined to be $\pm$11% at concentrations between 0.6-40 DPM/g at 95% confidence.

Ra-226 dating was completed using N40110 Determination of Radium-226 in Sediment, Soil, and Peat by Radon-222 Emanation. The MDL is dependent on the amount of sample analyzed. For a 60,000 second counting time the MDL at 95% confidence for 2 g of dry sample is 0.1 DPM/g and 0.5 g of dry sample is 0.5 DPM/g. The estimated uncertainty of measurement for this method at the Flett laboratory is approximately $\pm$12% at 95% confidence level (approximately 40,000 counts in 60,000 seconds).

Cs-137 dating was completed using N30120 Measurement of Gamma-Ray Emitting Radionuclides in Sediment/Soil Samples by Gamma Spectrometry Using HPGe Detectors. The MDL is 0.3 DPM/g for an 80,000 second counting period when measuring a 9 g of dry sample at a 95% confidence level. The method detection limit can be decreased to 0.1 DPM/g if 32 g of sample is used. The estimated uncertainty of this method has been determined to be $\pm$10% at 95% confidence for samples with activities between 0.5 and 20 DPM/g, counting time 80,000 seconds and sample weights ranging from 9 to 32 grams. Method uncertainty can increase to 85% for samples with activities near detection limit (0.1-0.3 DPM/g).

Dry bulk density is required on each sample in order to apply the CRS model to the data. A known volume of wet sediment was dried at about 50^{o}C and the dry weight of remaining sediment was determined. This is important because the Pb-210 is measured on dry samples and therefore calculations of how much Pb-210 is contained within in each ml of the original wet core material is needed. After the Pb-210 background was subtracted from each ml of sediment, the total amount of excess (atmospheric sourced) Pb-210 in the core as DPM/cm^2 was calculated. This total excess Pb-210 inventory, together with the subtotals upon which it is based, were used by the CRS model to calculate accumulation rates and ages for each core section.

A linear regression model was used to determine the age vs sediment depth relationship using unsupported Pb-210 activity and Cumulative dry weight (g/cm^2). When applying the linear regression model, it is assumed that the input of Pb-210 and the estimated sediment accumulation rate, based on the regression slope, are constant. This estimate of sediment accumulation rate was used to calibrate the CRS model.

A constant rate of Pb-210 supply (CRS) model was used to determine estimated accumulation rates using the sediment age at the bottom of each core section (in years) and the depth of each core section bottom (in cm). The CRS model assumes constant input of Pb-210 and a core that is long enough to include all measurable atmospheric source Pb-210, i.e. it contains a complete Pb-210 inventory. The CRS model is calibrated against the linear regression model to allow estimations of sediment ages through the core to be used to estimate sediment accumulation rates.

3. Results

3.1 Total Mercury

Total Mercury samples were collected from core sections at four sample locations as shown in Figure 1. Table 2 provides a summary of the dry and wet weight concentrations within each core and Figures 2 and 3 show the relative concentrations by depth for each site (dry wt and wet wt, respectively). As shown, dry weight concentrations are relatively consistent at each sample location through the core sections; however, concentrations are relatively greater in the outer sample locations in Lake Melville than Goose Bay. Wet weight concentrations are more variable along the sections of each core; showing generally increasing THg concentrations at deeper sections. A similar trend of increasing THg concentrations with distance from Goose Bay remains.

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Table 2: Summary of THg of each core section, Goose Bay and Lake Melville, May 2019. SE = Standard Error of the mean.

Sample Location	Sample Core Depth (cm)	THg (ng/g dry wt)	THg (ng/g wet wt)
Goose Bay (GB03)	0-1	12.3	4.97
	1-2	12.4	7.51
	2-3	13.1	9.10
	3-4	13.4	9.74
	4-5	12.2	8.91
	5-6	11.7	8.53
	6-7	12.5	8.80
	Mean (SE)	12.51 (0.215)	8.22 (0.599)
Lake Melville (LM02)	0-1	34.4	10.2
	1-2	34.7	14.0
	2-3	33.7	15.3
	3-4	30.4	14.7
	4-5	26.7	15.3
	5-6	26.8	16.0
	Mean (SE)	31.12 (1.52)	14.25 (0.855)
Lake Melville (LM03)	0-1	48.3	11.8
	1-2	49.6	16.5
	2-3	51.3	22.3
	3-4	48.2	18.9
	4-5	50.2	22.8
	5-6	52.4	23.3
	6-7	53.7	24.5
	7-8	54.0	24.4
	Mean (SE)	50.96 (0.802)	20.56 (1.591)
Lake Melville (LM04)	0-1	45.2	11.8
	1-2	47.0	15.4
	2-3	48.5	17.9
	3-4	50.7	20.1
	4-5	51.0	21.8
	5-6	51.8	25.0
	6-7	50.5	25.2
	7-8	51.2	25.8
	8-9	50.7	26.0
	9-10	49.3	25.2
	Mean (SE)	49.59 (0.667)	21.42 (1.580)

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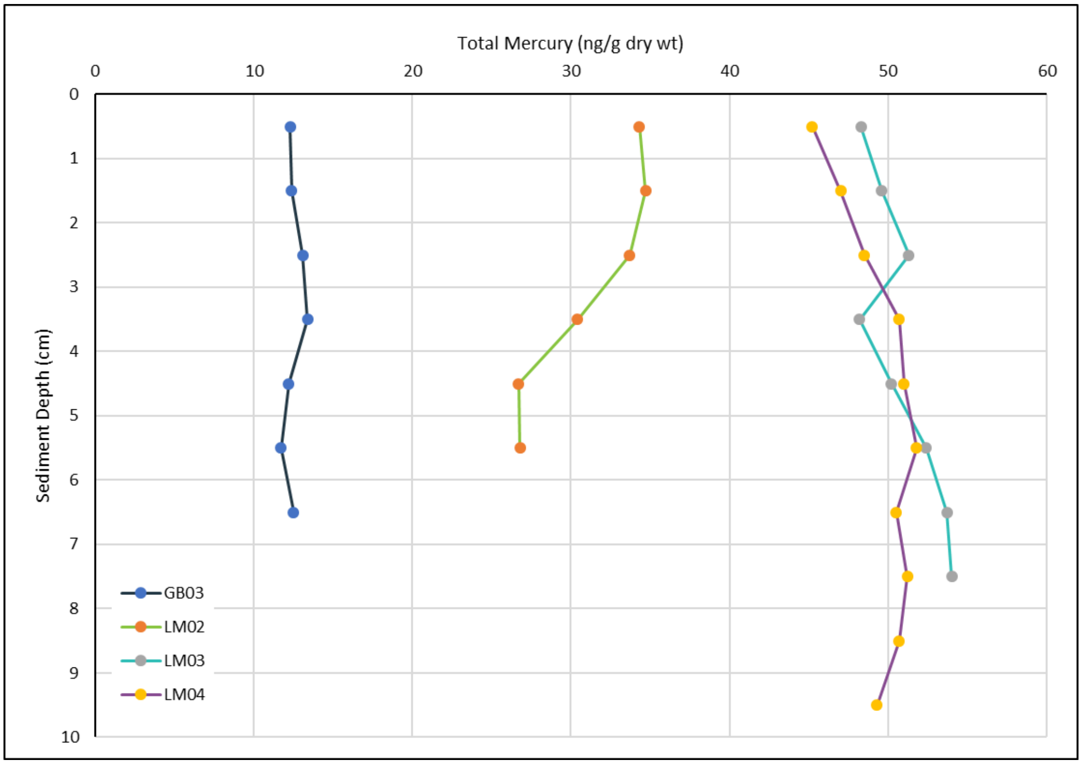

Figure 2: Total Mercury (ng/g) concentrations as dry weight

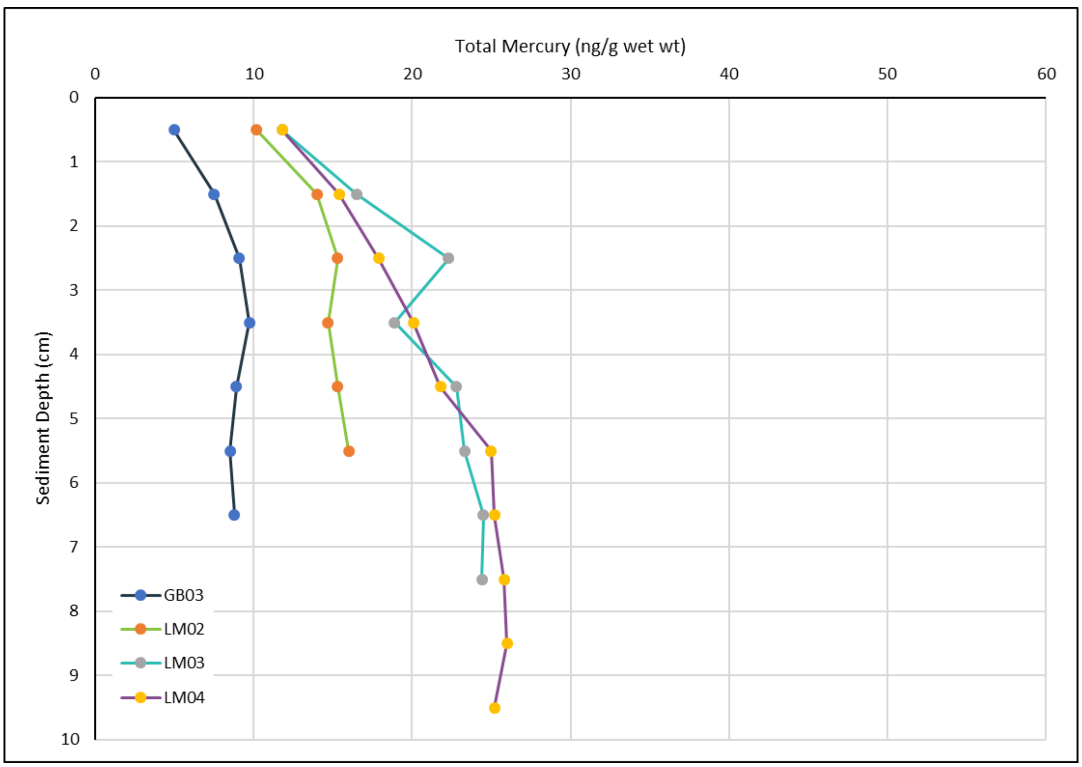

Figure 3: Total Mercury (ng/g) concentrations as wet weight

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3.2 Dating

Data and interpretation/modelling have been included on single XL97-2003 workbooks for each core, in a standardized format. Where possible, ages were assigned to the bottom of each section. Graphs of Pb-210 vs accumulated dry weight/cm^2, dry bulk density vs accumulated dry weight/cm^2, best fit regression of Pb-210 vs accumulated dry weight/cm^2, CRS and linear model age vs depth comparison, CRS sediment accumulation rates vs depth, CRS sediment accumulation rates vs age, and Cs-137 vs depth were included where possible. All raw data, calibrations and calculations are included in appended data outputs. Table 3 provides a summary of several core parameters.

Goose Bay (GB03)

The Pb-210 activity profile of this core shows an irregular but approximately exponential decrease as a function of depth. The maximum activity of 2.64 DPM/g observed in section 3 (depth 2 - 3 cm) is about 1.4 times the lowest activity of 1.92 DPM/g in section 6 (depth 5 - 6 cm). The Pb-210 activities in sections 1 and 2 (depth 0 - 2 cm) are slightly lower than the Pb-210 activity in section 3, and this probably represents increasing sediment accumulation rates, and/or physical mixing, and/or diffusion of Pb-210 across a redox gradient, and/or incomplete diagenesis of surface sediment, and/or incomplete ingrowth of the Po-210, granddaughter of Pb-210, actually being measured.

The dry bulk densities gradually increase with depth from 0.555 g/cm^3 at section 1 (depth 0 - 1 cm) to 1.368 g/cm^3 at section 4 (depth 3 - 4 cm) and then decrease to 1.295 g/cm^3 at the bottom section (depth 6 - 7 cm) (Table 3).

Ra-226 was measured at 0.81, 0.87 and 0.70 DPM/g in sections 2, 4 and 7, respectively. Net unsupported Pb-210 was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value, unless noted otherwise. The Pb-210 activity in section 7 was significantly higher than the Ra-226 activity measured in the same section, indicating that the background level of Pb-210 has not been achieved in this core.

The Cs-137 activities were low, varying between 0.18 - 0.38 DPM/g throughout the core.

The linear regression model predicts (R^2 = 0.6942) an average sediment accumulation rate of 0.5076 g/cm^2/yr when the unsupported Pb-210 activity was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value. The age at the bottom of any core section can be estimated by dividing the cumulative dry weight/cm^2 by the accumulation rate. The age estimate at the bottom of each section is shown in Table 3.

The CRS model assumes constant input of Pb-210 and a core that is long enough to include all measurable atmospheric source Pb-210, i.e. it contains a complete Pb-210 inventory. Since the second assumption is not satisfied in this core (background has not been achieved), it is not normally possible to apply the CRS model. However, in this core it is possible to calibrate the CRS model against the linear regression model, and therefore allow the CRS model to be used over the entire core. The total atmospheric Pb-210 inventory (DPM/cm^2), required in the CRS model calculation, has been chosen (30.990 DPM/cm^2) such that the CRS model predicted exactly the same average sediment accumulation rate (0.5076 g/cm^2/yr) as the linear regression model. With the CRS model calibrated, it has been used to calculate ages for the entire core.

The average sediment accumulation rate, from core surface to the extrapolated bottom depth of any section, was calculated by dividing the cumulative dry mass at the bottom of the extrapolated section by the calculated age at that depth (Table 3). Figures 4 and 5 plot age vs. depth and sediment accumulation rate vs. age, respectively.

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Table 3: Summary of sediment core parameters based on Pb-210, Ra226, and Cs-137 analyses

Sample Location	Sample Core Depth (cm)	Dry Bulk Density (Dry wt/Wet wt vol) (g/cm^3)	Linear Regression Age at Bottom of Extrapolated Section (years)	CRS Age at Bottom of Extrapolated Section (years)	CRS Sediment Accumulation Rate ($g/cm^2/yr$)
Goose Bay (GB03)	0-1	0.555	1.1	0.9	0.6104
	1-2	0.987	3.0	2.8	0.5197
	2-3	1.264	5.5	5.5	0.4782
	3-4	1.368	8.2	8.1	0.5237
	4-5	1.363	10.9	10.8	0.4910
	5-6	1.363	13.6	13.4	0.5406
	6-7	1.295	16.1	16.1	0.4653
	Average				**0.5076**
Lake Melville (LM02)	0-1	0.366	3.7	3.5	0.1038
	1-2	0.543	9.2	8.7	0.1056
	2-3	0.641	15.7	15.7	0.0908
	3-4	0.709	22.9	23.4	0.0929
	4-5	0.906	32.0	31.6	0.1096
	5-6	0.967	41.8	41.8	0.0946
	Average				**0.0988**
Lake Melville (LM03)	0-1	0.289	1.6	1.6	0.1850
	1-2	0.425	4.0	4.0	0.1728
	2-3	0.604	7.4	7.6	0.1670
	3-4	0.523	10.4	10.3	0.1967
	4-5	0.638	14.0	13.8	0.1840
	5-6	0.621	17.5	17.2	0.1833
	6-7	0.644	21.1	21.0	0.1672
	7-8	0.615	24.5	24.5	0.1740
	Average				**0.1776**
Lake Melville (LM04)	0-1	0.312	-	-	-
	1-2	0.418	-	-	-
	2-3	0.486	-	-	-
	3-4	0.534	-	-	-
	4-5	0.591	-	-	-
	5-6	0.699	-	-	-
	6-7	0.735	-	-	-
	7-8	0.742	-	-	-
	8-9	0.761	-	-	-
	9-10	0.759	-	-	-
	Average				**0.1054**

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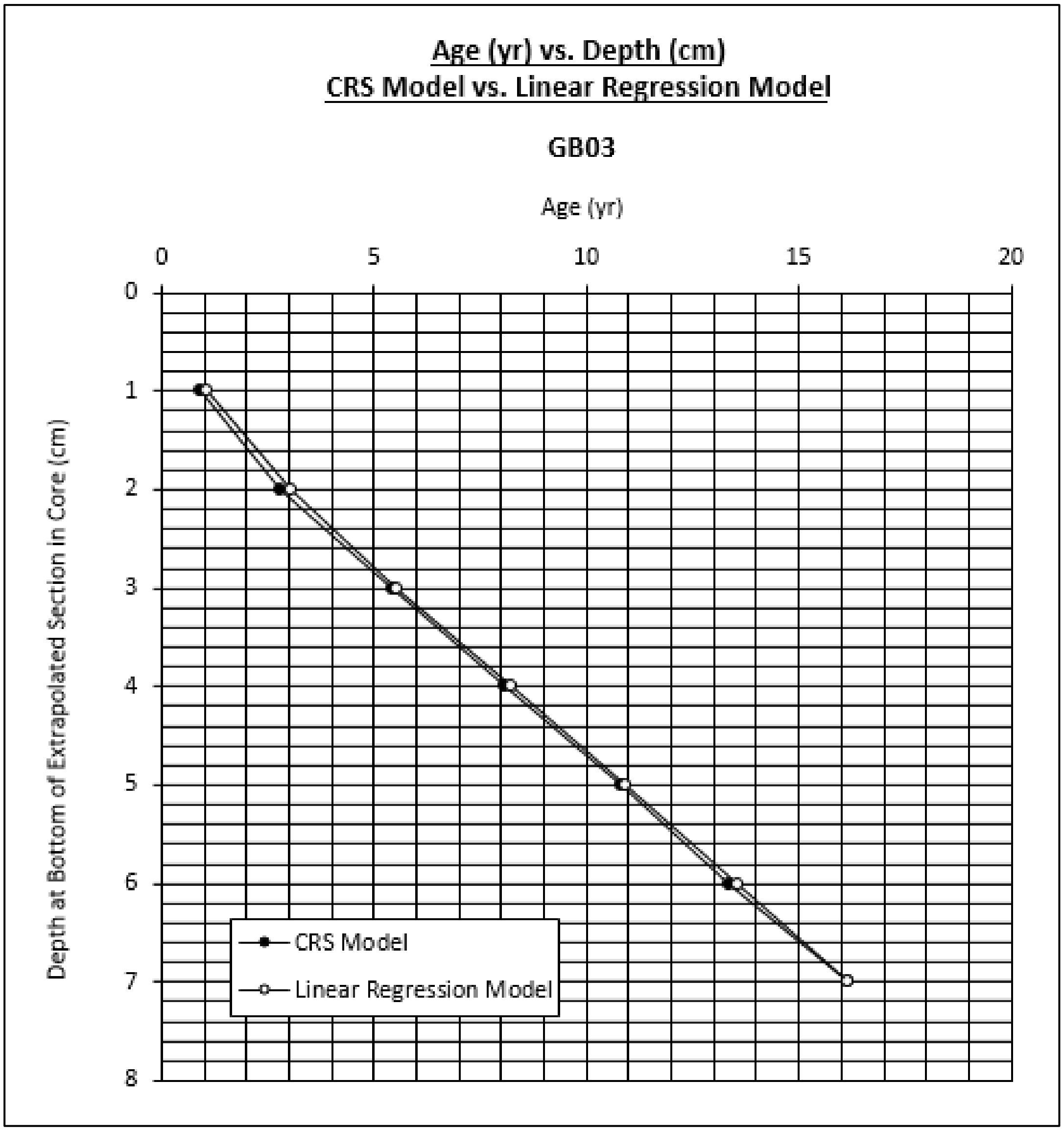

Figure 4: Age vs depth regression model results, GB03

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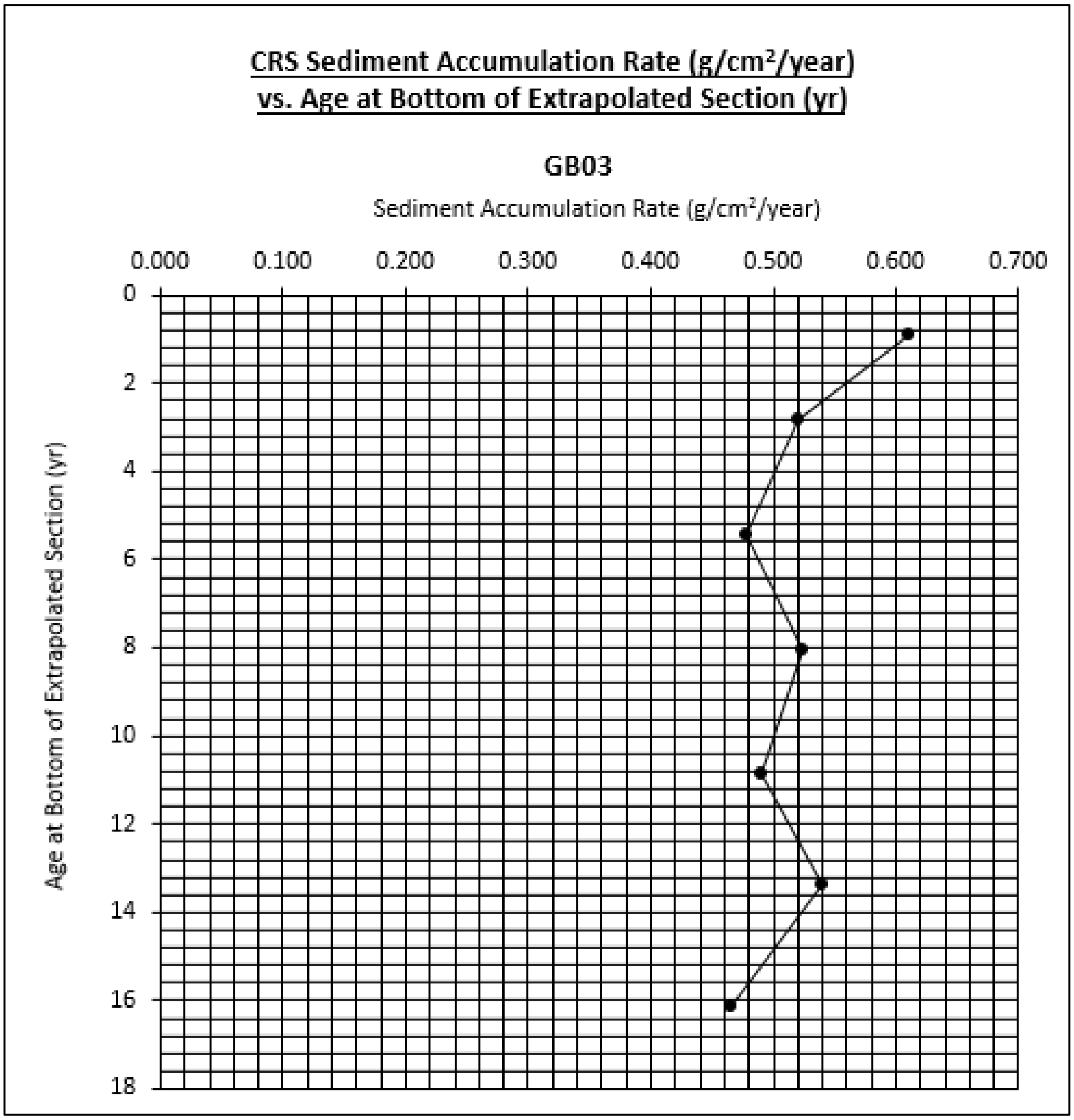

Figure 5: Sediment Accumulation Rate vs Sediment Age, GB03

Conclusion

The sediment accumulation rates within the Goose Bay sample varied between 0.4633 and 0.6104 g/cm^2/yr throughout the core (by the CRS model and the shape of Pb-210 activity profile).

The constantly low Cs-137 activities suggests that the majority of the Cs-137 is probably from external erosion sources (soils or sediments contaminated with bomb testing radionuclides) rather than direct deposition from the atmosphere. Based upon the shape of Cs-137 profile, it is likely that the 1966 maximum Cs-137 terrestrial inventory could be attained below 7 cm depth, suggesting that all sections (0 - 7 cm) are less than 53 years old (post 1966). The CRS model indicates an age of 16.1 yr at the bottom of the core (7 cm depth), an age compatible with the presence of Cs-137.

Over the entire core length, the average sediment accumulation rate estimated by the CRS model has been forced to exactly coincide with the linear regression estimate of 0.5076 g/cm^2/yr. Although the CRS calculated ages depend

upon the results of the linear regression model, the CRS model is to be preferred because it should provide accurate age predictions at the bottom of each section even though the sediment accumulation rate is changing with time.

Overall, the analytical quality of radioisotope data (based upon the recovery of spike, the recovery of CRM, the results of repeat analyses and blanks) is considered good.

Lake Melville (LM02)

The Pb-210 activity profile of this core shows an irregular but approximately exponential decrease as a function of depth. The maximum activity of 6.19 DPM/g observed in section 1 (depth 0 - 1 cm) is about 2.2 times the lowest activity of 2.83 DPM/g in section 6 (depth 5 - 6 cm).

The dry bulk densities gradually increase with depth from 0.366 g/cm3 at section 1 (depth 0 - 1 cm) to 0.967 g/cm^3 at section 6 (depth 5 - 6 cm).

Ra-226 was measured at 1.06, 1.12 and 0.92 DPM/g in sections 1, 4 and 6, respectively. Net unsupported Pb-210 was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value, unless noted otherwise. The Pb-210 activity in section 6 is significantly higher than the Ra-226 activity measured in the same section, indicating that the background level of Pb-210 has not been achieved in this core.

The Cs-137 activities are low, varying between 0.34 - 0.59 DPM/g throughout the core.

The linear regression model predicts (R^2 = 0.9639) an average sediment accumulation rate of 0.0988 g/cm^2/yr when the unsupported Pb-210 activity was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value. The age at the bottom of any core section can be estimated by dividing the cumulative dry weight/cm^2 by the accumulation rate. The age estimate at the bottom of each section is shown in Table 3.

The CRS model assumes constant input of Pb-210 and a core that is long enough to include all measurable atmospheric source Pb-210, i.e. it contains a complete Pb-210 inventory. Since the second assumption is not satisfied in this core (background has not been achieved), it is not normally possible to apply the CRS model. However, in this core it is possible to calibrate the CRS model against the linear regression model, and therefore allow the CRS model to be used over the entire core. The total atmospheric Pb-210 inventory (DPM/cm^2), required in the CRS model calculation, has been chosen (18.120 DPM/cm^2) such that the CRS model predicted exactly the same average sediment accumulation rate (0.0988 g/cm^2/yr) as the linear regression model. With the CRS model calibrated, it has been used to calculate ages for the entire core.

The average sediment accumulation rate, from core surface to the extrapolated bottom depth of any section, was calculated by dividing the cumulative dry mass at the bottom of the extrapolated section by the calculated age at that depth (Table 3). Figures 6 and 7 plot age vs. depth and sediment accumulation rate vs. age, respectively.

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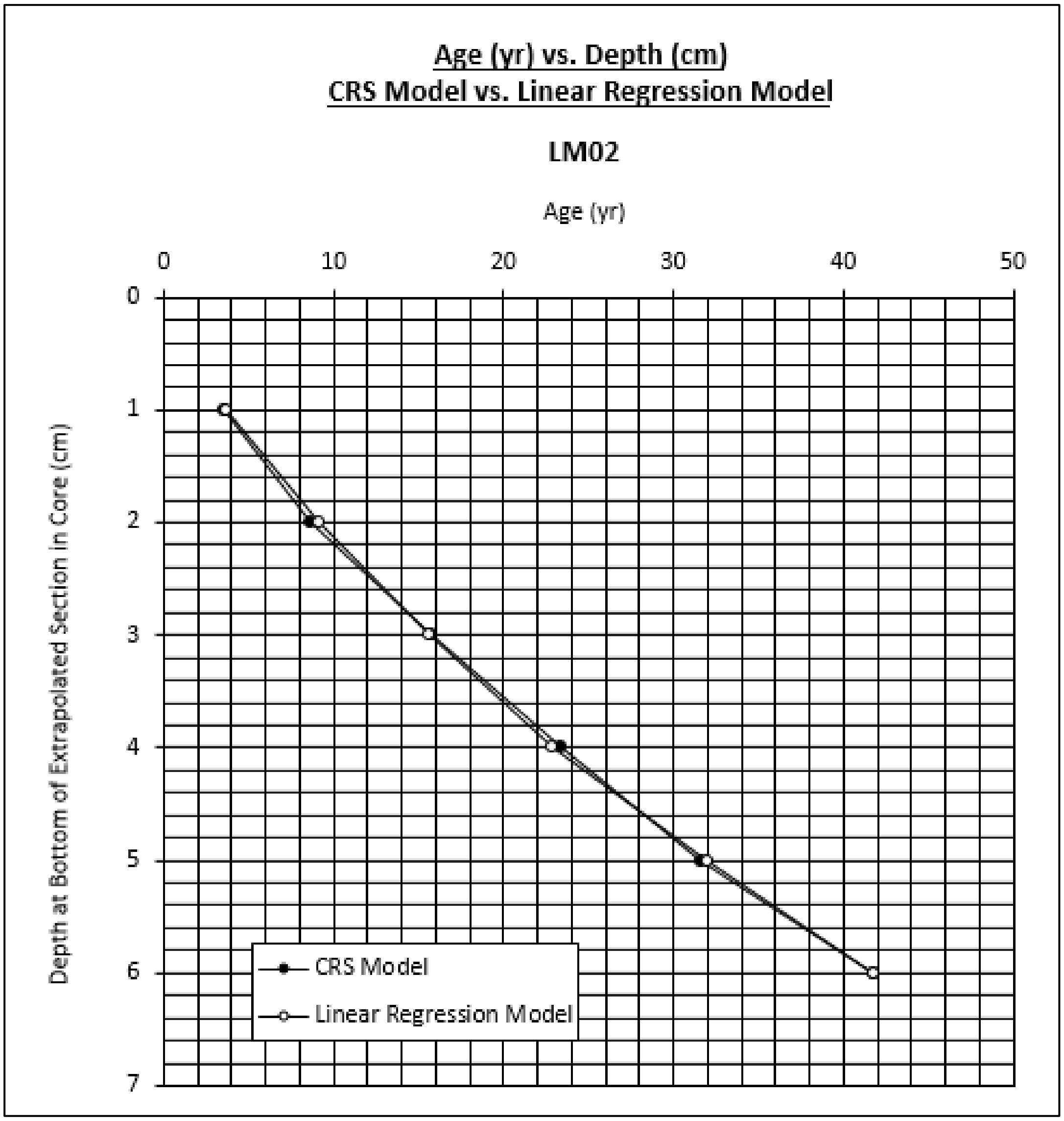

Figure 6: Sediment Accumulation Rate vs Sediment Age, LM02

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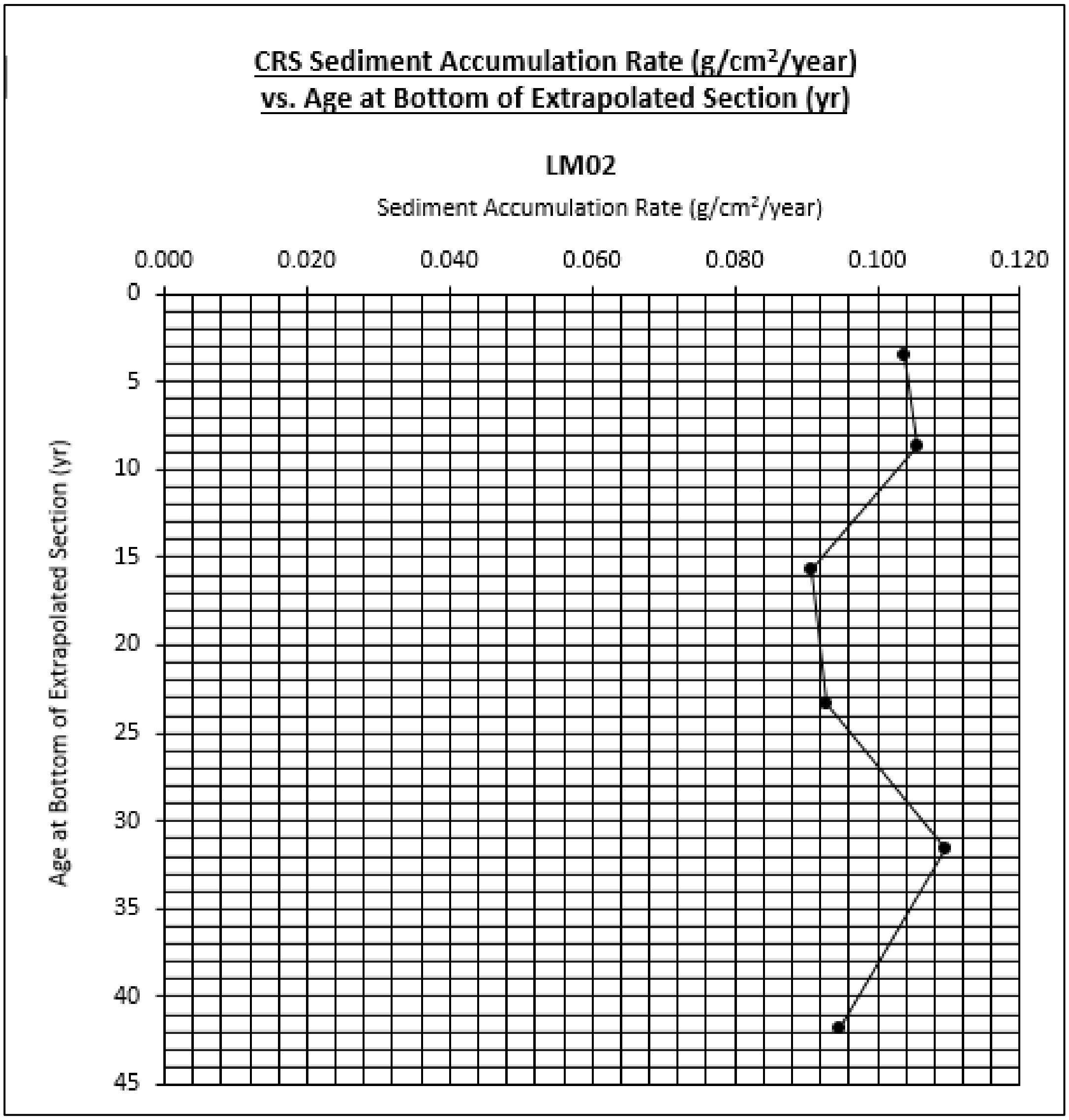

Figure 7: Age vs depth regression model results, LM02

Conclusion

The sediment accumulation rates vary between 0.0908 g/cm^2/yr and 0.1096 g/cm^2/yr throughout the core (by the CRS model and the shape of Pb-210 activity profile).

The constantly low Cs-137 activities suggests that the majority of the Cs-137 is probably from external erosion sources (soils or sediments contaminated with bomb testing radionuclides) rather than direct deposition from the atmosphere. Based upon the shape of Cs-137 profile, it is likely that the 1966 maximum Cs-137 terrestrial inventory could be attained below 6 depth, suggesting that all sections (0 - 6 cm) are less than 53 years old (post 1966). The CRS model indicates an age of 41.8 yr at the bottom of the core (7 cm depth), an age compatible with the presence of Cs-137.

Over the entire core length, the average sediment accumulation rate estimated by the CRS model has been forced to exactly coincide with the linear regression estimate of 0.0988 g/cm^2/yr. Although the CRS calculated ages depend

upon the results of the linear regression model, the CRS model is to be preferred because it should provide accurate age predictions at the bottom of each section even though the sediment accumulation rate is changing with time.

Overall, the analytical quality of radioisotope data (based upon the recovery of spike, the recovery of CRM, the results of repeat analyses and blanks) is considered good.

Lake Melville (LM03)

The Pb-210 activity profile of this core shows an irregular but approximately exponential decrease as a function of depth. The maximum activity of 8.45 DPM/g observed in section 2 (depth 1 - 2 cm) is about 1.5 times the lowest activity of 5.60 DPM/g in section 8 (depth 7 - 8 cm). The Pb-210 activity in sections 1 (depth 0 - 1 cm) are slightly lower than the Pb-210 activity in section 2, and this probably represents increasing sediment accumulation rates, and/or physical mixing, and/or diffusion of Pb-210 across a redox gradient, and/or incomplete diagenesis of surface sediment, and/or incomplete ingrowth of the Po-210, granddaughter of Pb-210, actually being measured.

The dry bulk densities gradually increase with depth from 0.289 g/cm3 at section 1 (depth 0 - 1 cm) to 0.604 g/cm^3 at section 3 (depth 2 - 3 cm), then decrease to 0.523 g/cm3 at section 4 (depth 3 - 4 cm) and vary between 0.615 g/cm^3 and 0.644 g/cm^3 below 4 cm depth.

Ra-226 was measured at 2.19, 2.39 and 2.26 DPM/g in sections 1, 3 and 8, respectively. Net unsupported Pb-210 was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value, unless noted otherwise. The Pb-210 activity in section 8 is significantly higher than the Ra-226 activity measured in the same section, indicating that the background level of Pb-210 has not been achieved in this core.

The Cs-137 activities are low, varying between 0.54 - 0.79 DPM/g in the upper 7 cm of the core. No detectable Cs-137 was observed in sample 3 - 4 cm duplicate and sample 7 - 8 cm, which could result from the small sample size (<3 g dry weight) available.

The linear regression model predicts ($R^2 = 0.9449$) an average sediment accumulation rate of 0.1776 $g/cm^2/yr$ when the unsupported Pb-210 activity was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value. The age at the bottom of any core section can be estimated by dividing the cumulative dry weight/cm^2 by the accumulation rate. The age estimate at the bottom of each section is shown in Table 3.

The CRS model assumes constant input of Pb-210 and a core that is long enough to include measurable atmospheric source Pb-210, i.e. it contains a complete Pb-210 inventory. Since the second assumption is not satisfied in this core (background has not been achieved), it is not normally possible to apply the CRS model. However, in this core it is possible to calibrate the CRS model against the linear regression model, and therefore allow the CRS model to be used over the entire core. The total atmospheric Pb-210 inventory (DPM/cm^2), required in the CRS model calculation, has been chosen (37.950 DPM/cm^2) such that the CRS model predicted exactly the same average sediment accumulation rate (0.1776 $g/cm^2/yr$) as the linear regression model. With the CRS model calibrated, it has been used to calculate ages for the entire core.

The average sediment accumulation rate, from core surface to the extrapolated bottom depth of any section, can be calculated by dividing the cumulative dry mass at the bottom of the extrapolated section by the calculated age at that depth (Table 3). Figures 8 and 9 plot age vs. depth and sediment accumulation rate vs. age, respectively.

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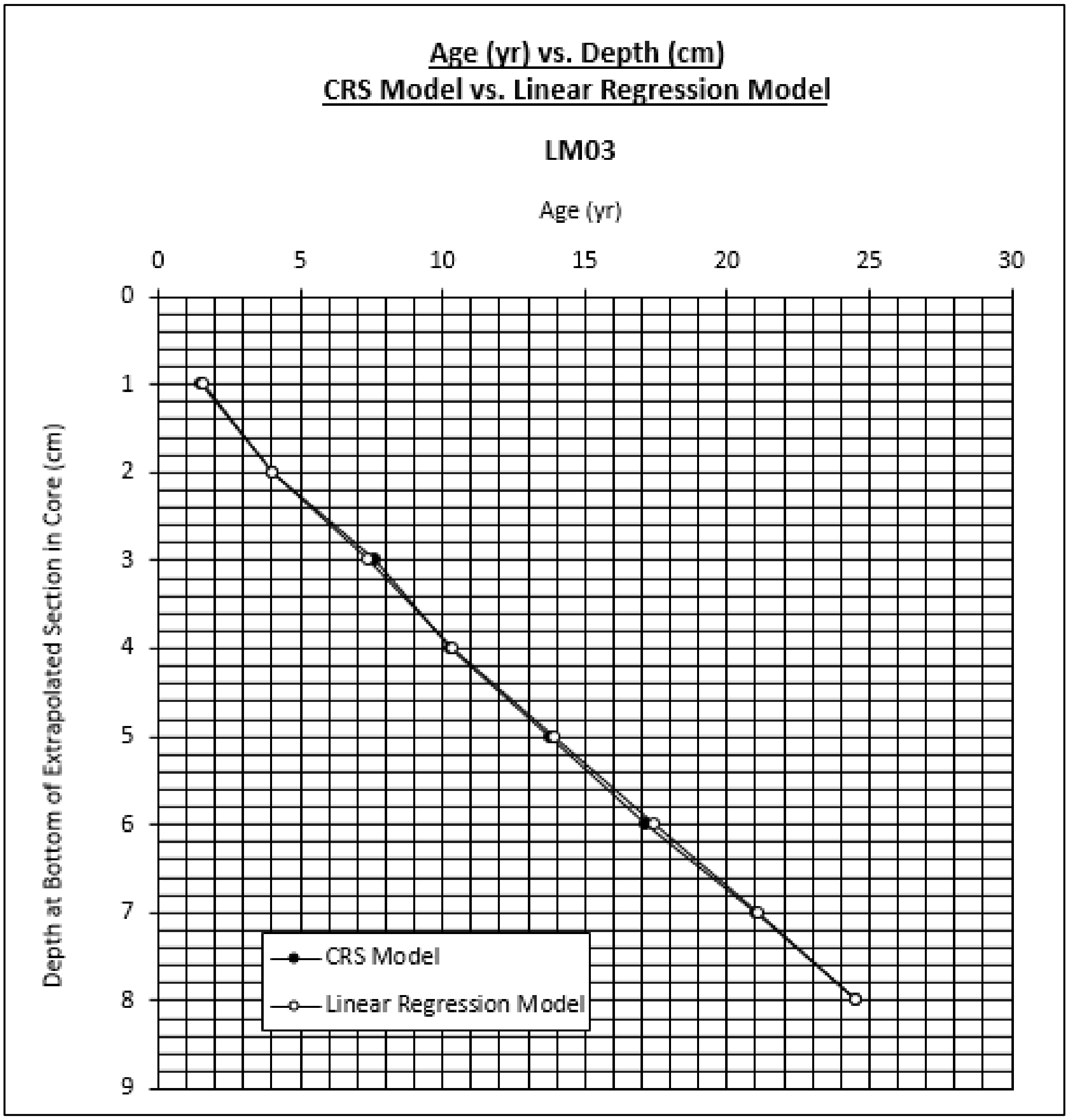

Figure 8: Sediment Accumulation Rate vs Sediment Age, LM03

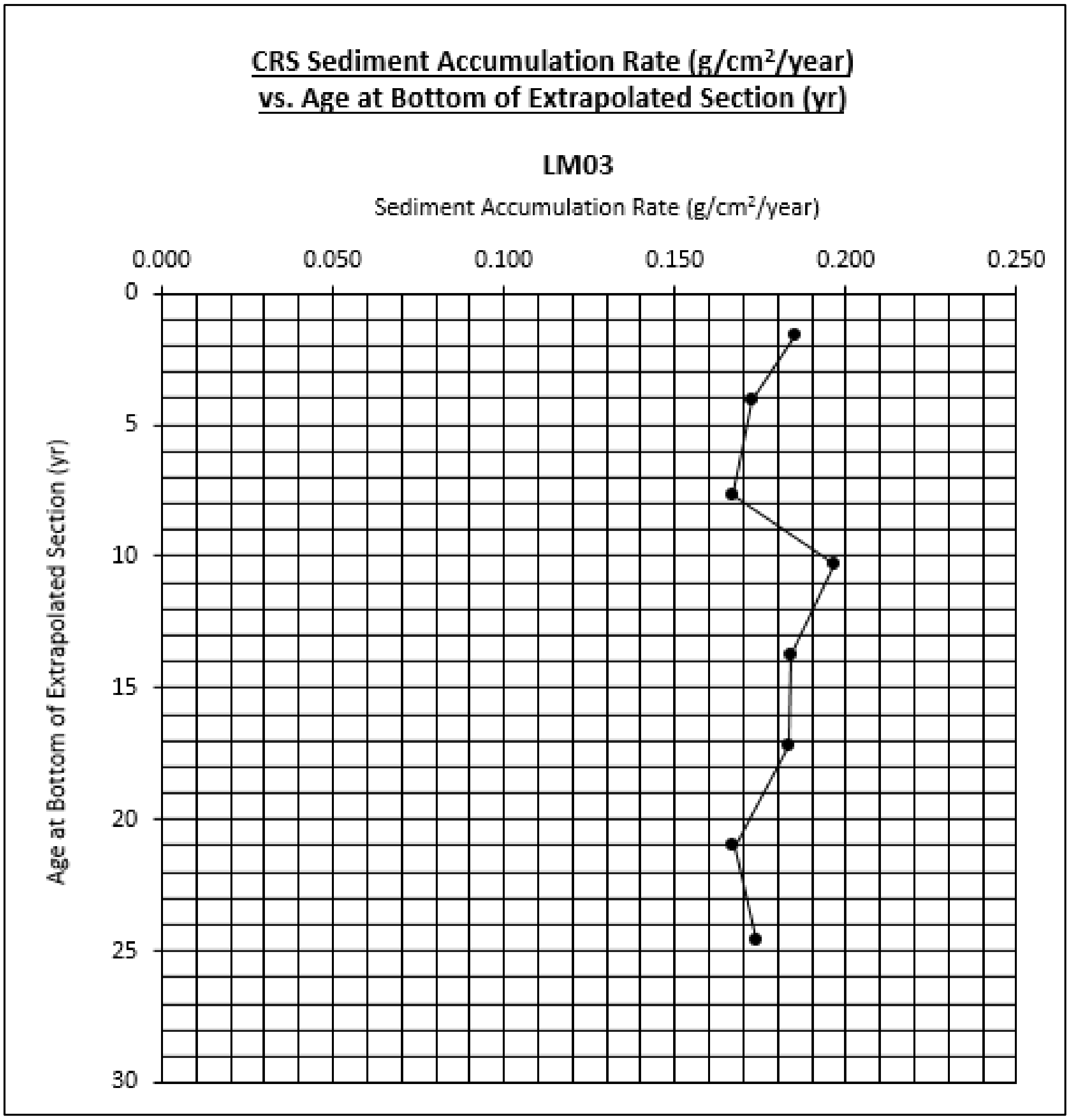

Figure 9: Age vs depth regression model results, LM03

Conclusion

The sediment accumulation rates vary between 0.1670 and 0.1967 g/cm^2/yr throughout the core (by the CRS model and the shape of Pb-210 activity profile).

The constantly low Cs-137 activities suggests that the majority of the Cs-137 is probably from external erosion sources (soils or sediments contaminated with bomb testing radionuclides) rather than direct deposition from the atmosphere. Based upon the shape of Cs-137 profile, it is likely that the 1966 maximum Cs-137 terrestrial inventory could be attained below 8 depth, suggesting that all sections (0 - 8 cm) are less than 53 years old (post 1966). The CRS model indicates an age of 24.5 yr at the bottom of the core (8 cm depth), an age compatible with the presence of Cs-137.

Over the entire core length, the average sediment accumulation rate estimated by the CRS model has been forced to exactly coincide with the linear regression estimate of 0.1776 g/cm^2/yr. Although the CRS calculated ages depend

upon the results of the linear regression model, the CRS model is to be preferred because it should provide accurate age predictions at the bottom of each section even though the sediment accumulation rate is changing with time.

Overall, the analytical quality of radioisotope data (based upon the recovery of spike, the recovery of CRM, the results of repeat analyses and blanks) is considered good.

Lake Melville (LM04)

The Pb-210 activity profile of this core shows an irregular but approximately exponential decrease as a function of depth. The maximum activity of 9.08 DPM/g observed in section 1 (depth 0 - 1 cm) is about 2.5 times the lowest activity of 3.61 DPM/g in section 10 (depth 9 - 10 cm).

The dry bulk densities gradually increase with depth from 0.312 g/cm^3 at section 1 (depth 0 - 1 cm) to 0.761 g/cm^3 at section 9 (depth 8 - 9 cm) (Table 3).

Ra-226 was measured in 6 sections and the activities significantly decrease from 2.95 DPM/g in the surface section (depth 0 - 1 cm) to 1.18 DPM/g in the bottom section (depth 9 - 10 cm) (Pages 7 - 14). Net unsupported Pb-210 was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value, unless noted otherwise. The Pb-210 activity in section 10 is significantly higher than the Ra-226 activity measured in the same section, indicating that the background level of Pb-210 has not been achieved in this core.

The Cs-137 activities generally increase with depth from 0.39 DPM/g in the surface section (depth 0 - 1 cm) to 1.08 DPM/g in the bottom section (depth 9 - 10 cm).

When applying the linear regression model, it is assumed that the input of Pb-210 and the sediment accumulation rate are constant. Although some variation in the sediment accumulation rate is apparent, the linear regression model was applied to the core interval of 5 - 10 cm, because it appears that the average sediment accumulation rate in this core interval will be reasonably estimated. The linear regression model predicts (R^2 = 0.9868) an average sediment accumulation rate of 0.1054 g/cm^2/yr when the unsupported Pb-210 activity was calculated by subtracting the nearest neighbouring Ra-226 measurement from each total Pb-210 value. The number of years included in the 5 - 10 cm core interval can be estimated by dividing the cumulative dry weight/cm^2 by the accumulation rate, which is calculated as (6.035-2.340) / 0.1054 = 35.1 yr.

The CRS model assumes constant input of Pb-210 and a core that is long enough to include all of the measurable atmospheric source Pb-210, i.e. it contains a complete Pb-210 inventory. Since the second assumption is not satisfied in this core (background has not been achieved), it is not normally possible to apply the CRS model. Additionally, based upon the significant changes in Ra-226 activities in the upper 5 cm of the core, it is assumed that the core interval of 0 - 5 cm consisted of modern, different sourced sediment, and could be the result of a short duration depositional event (or events) that may have imported foreign excess Pb-210 inventory. This atypical input of sediment will cause the CRS model to fail because the assumption of a constant Pb-210 input rate is no longer valid.

Conclusion

In the core interval of 5 - 10 cm, the average sediment accumulation rate is 0.1054 g/cm^2/yr, estimated by the linear regression model.

The shape of Cs-137 profile suggests that the majority of the Cs-137 is probably from external erosion sources (soils or sediments contaminated with bomb testing radionuclides) rather than direct deposition from the atmosphere. Based upon the shape of Cs-137 profile, it is likely that the 1966 maximum Cs-137 terrestrial inventory could be attained below 10 depth, suggesting that all sections (0 - 10 cm) are less than 53 years old (post 1966).

Estimated by the linear regression model, the 5 - 10 cm core interval contains about 35 years of sediment. The age at 5 cm depth cannot be calculated by either the linear regression model or the CRS model, due to high uncertainty in the sedimentation processes in the upper 5 cm of the core. However, at 5 cm depth the age of the sediment is probably less than 53 - 35 = 18 years, in order to agree with the conclusion that all sections (0 - 10 cm) were deposited post 1966.

Overall, the analytical quality of radioisotope data (based upon the recovery of spike, the recovery of CRM, the results of repeat analyses and blanks) is considered good.

4. CLOSURE

If you have any questions, or require any further information, please feel free to contact me at your convenience.

Yours sincerely,

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