Refining the marine reservoir effect in the Northwest North Atlantic

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Abstract

This research examines whether ^{14}C dating of pollen grains can be used as an alternative dating method for marine sediments and if the pollen ages can be used to refine the value of the Marine Reservoir Effect (MRE) applied to marine carbonates from cores retrieved from along the East Canadian Margin (Newfoundland and Scotian Shelves). Precise dating is critically important to situate abrupt climate events, such as the onsets and conclusions of the 8.2 ka cooling event, Younger Dryas, and Preboreal Oscillation and sediments thought relevant to these events have been used in my study.

Pollen was extracted from ocean and wetland sediments cored from the eastern Canadian margin, James Bay region and Maritime Provinces of Canada and ^{14}C dated using Accelerator Mass Spectrometry (AMS). Pollen dates from ocean sediments were compared with marine carbonate (mollusk shells or foraminifera) dates from the same core levels for which reworking has been excluded. The validity of the core dates was assessed via correlations with cores from other studies. Pollen samples from three tidal wetlands were taken from levels dated with ^{137}Cs and ^{210}Pb profiles. Ages of pollen from two additional wetlands were compared with ^{14}C dates of botanical macrofossils.

Most pollen dates vary from ^{14}C dates based on macrofossils or carbonates, with age differences typically exceeding 250 years and over 3000 years in one instance. In some samples pollen ages were younger than the corresponding carbonate, as expected, since pollen ages should not be affected by the MRE. These differences in age between these MRE affected carbonates and the non MRE affected pollen, suggest that a MRE and the associated reservoir age could alter our understanding of the climatic timeline for

the eastern Canadian Margin. However, these new reservoir ages have yet to be validated with the stratigraphic interpretations of the cores with which they are associated.

In some cores, however, pollen dates show age reversals. Significant proportions of reworked pollen grains in ocean and wetland samples are associated with pollen dates that are too old. Prolonged core storage could result in pollen ^{14}C ages that are too young, possibly because of bacterial growth but more work is needed to verify this hypothesis. Despite the problems encountered, some pollen dates are consistent with other ^{14}C dates from the same core levels, suggesting this dating method can work. At present however, more work is needed to understand the conflicting results obtained.

Résumé

Cette recherche vise à determiner si la datation par carbone 14 des grains de pollen peut être utilisée comme méthode alternative de datation des sédiments marins et si l'âge du pollen peut être utilisé pour raffiner la correction due à l'effet réservoir marin (Marine Reservoir Effect) appliquée aux carbonates marins des carottes prélevée le long de la marge de l'est canadien (plateaux continental de Terre-neuve et Néo-Écossais). Une datation précise est d'une grande importance afin de situer dans le temps des évènements climatiques soudains comme le début et la fin de la période de refroidissement du 8.2 ka, l'oscillation du Dryas récent et l'oscillation du Préboréal et les sédiments correspondant à ces évènements ont été utilisés dans cette étude.

Du pollen a été extrait de sédiments océaniques et de zones humides prélevés sur la marge est-canadienne, dans la région de la Baie James et dans les provinces maritimes du Canada et datés au ^{14}C en utilisant la méthode d`accélération de particules et le spectromètre de mass « Accelerator Mass Spectrometry (AMS) ». Les dates basées sur le pollen des sédiments marins ont été comparées avec celles obtenues sur des carbonates marins (coquilles de mollusques et foraminifères) provenant des mêmes niveaux dans les carottes, pour lesquelles le remaniement a été exclu. La validité des dates des carottes a été évaluée via des corrélations avec des carottes provenant d`autres études, également datées. Des échantillons de pollen de trois zones humides de marrée ont été pris aux niveaux qui ont été préalablement datés avec des profils de ^{137}Cs et ^{210}Pb. Lesâges du pollen de deux autres zones humides ont été comparés avec les âges ^{14}C de macrorestes botaniques.

La plupart des dates ^{14}C basées sur le pollen diffèrent de celles basées sur les macrorestes ou les carbonates, avec des différences d'âge de plus de 250 ans, et même de 3000 ans dans un cas. Dans quelques échantillons, les âges basées sur le pollen sont plus jeunes que ceux des carbonates du même niveau, tel que prévu. Ces différences entre les âges des carbonates, qui sont affectés par l'effet réservoir marin et les âges du pollen, qui ne sont pas par l'effet réservoir marin, semble indiquer un effet réservoir marin, et la correction d`âge réservoir associée à cet effet pourrait altérer notre compréhension de la chronologie climatique de la marge continentale de l`est de Canada. Cependant, ces nouveaux âges réservoir n'ont pas encore été validés avec les interprétations stratigraphiques des carottes avec lequelles ils sont associés.

Toutefois, dans certaines carottes, les dates du pollen montrent des inversions d'âge. Des proportions significatives des grains de pollen retravaillés, dans les milieux marins et les zones humides, ont été associées avec des dates polliniques trop âgées. L'entreposage prolongé des carottes pourrait résulter en des dates ^{14}C trop jeunes, possiblement dû à la croissance de bactéries mais plus de recherche est nécessaire pour vérifier cette hypothèse. Malgré les difficultés rencontrées, quelques dates basées sur le pollen sont consistentes avec les autres dates ^{14}C du même niveau dans la carotte, suggérant que cette méthode de datation peut fonctionner. Pour l'instant, plus de recherche est nécessaire pour comprendre les résultats contradictoires obtenus.

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Chapter 1: Background and Literature Review

The importance of dating in paleoclimatic research

Paleoclimatic research requires sound chronological references (Brown et al. 1989; Eirıksson et al. 2004) to reconstruct event succession or to identify the cause of a climatic event such as the 8.2 ka cooling event, Younger Dryas, and the Preboreal Oscillation. It also is necessary to constrain the time period during which climatic, environmental or ecological events occurred. For Quaternary research one of the most common methods to obtain dates from ocean sediment samples is radiocarbon dating of carbonate from shells or foraminifera tests. While radiocarbon dates obtained with this method are generally valid; they must be corrected for the Marine Reservoir Effect (MRE), which varies with location and time. Reference data for the MRE is sparse, especially in the western North Atlantic. My research examines whether radiocarbon dating of pollen grains, which should not be affected by the MRE, could be used as an alternative dating method for marine sediments and if so, to determine more precisely the MRE along the East Canadian Margin (Newfoundland and Scotian Shelves) for the time periods mentioned above. To do this, sediment cores which had been previously dated using shells or foraminifera were subsampled at the same or similar levels from which the carbonates were obtained. Pollen grains were extracted from the samples, dated, and then compared to the dates derived from the carbonate assuming that the difference in age would equal the MRE for that location and time.

The basics of radiocarbon (^{14}C) dating

Bowman (1990) has reviewed the multiple methods used in ^{14}C dating, and compared their accuracy and efficiency in a book upon which much of this summary is

based. Methods include gas counting, liquid scintillation, and most recently, accelerated mass spectrometry (AMS) which was developed in the late 1970s. The differences between the gas counting and liquid scintillation methods, also known as conventional methods, and AMS are twofold. The conventional ^{14}C dating techniques derive dates from samples by measuring the beta particles that are released during the decay process of ^{14}C isotopes. While this method is quite accurate with margins of error of ±1% or ±80 years, its efficiency is quite low. The conventional methods only utilize a fraction of the ^{14}C atoms that are actually available within the sample during a 24 hour counting period. As such, these methods require either large quantities of sample carbon or extended counting periods to ensure that the appropriate margin of error is met. On the other hand, AMS dating derives ^{14}C dates with the same margin of error as conventional methods, but with shorter counting periods and a relatively small sample size (roughly 1,000 times smaller). Instead of measuring the released beta particles from the ^{14}C isotopes, AMS dating directly measures the ratio of ^{14}C isotopes relative to stable carbon isotopes (^{12}C and ^{13}C). This ratio is compared with that of a standard. Since ^{12}C and ^{13}C do not decay, their concentrations should not change; hence differences in the ^{14}C ratio can be used to determine the amount of ^{14}C decay within the sample.

One of the principal assumptions of ^{14}C dating is that the concentrations of the three carbon isotopes within the sample material are in relative equilibrium with those of the atmosphere at the time that the sample was deposited or ceased to live (Bowman 1990). This is because all carbon-based organisms will incorporate the three carbon isotopes into their carbon structure initially via photosynthesis and the uptake of CO_2 or by ingestion by consumer organisms (Bowman 1990). Upon death, the organism will

begin to lose ^{14}C through radioactive decay. The 'half-life' is the interval of time in which half the isotope decays. For ^{14}C, the half-life is ~5,568 years (Taylor 1987; Bowman 1990; Geyh and Schleicher 1990; Currie 2004).

However, concentrations of carbon isotopes throughout the biosphere are not constant (Currie 2004). The uptake of carbon isotopes does not occur in equilibrium with atmospheric concentrations during photosynthesis. Uptake is affected by the fixation mechanisms used by plant species and kinetic fractionation that causes ^{12}C to be more readily integrated than the heavier ^{13}C and even heavier ^{14}C (Bowman 1990; Geyh and Schleicher 1990; Coltrain 2005). While corrections have been developed to account for fractionation within plants, these discrepancies bring to question other environmental processes that cause fractionation of carbon isotopes. One of those processes in the marine environment is the MRE which affects radiocarbon dates based on carbonates secreted by marine organisms such as foraminifera and mollusks.

Another important point, as it relates to this study, is that it is assumed that the production and concentration of ^{14}C has remained constant over time in the atmosphere. However, the rate of production and concentration of the ^{14}C in the atmosphere has changed over time (Maier-Reimer and Hasselmann 1987; Stuiver and Braziunas 1989; Bowman 1990; Southon et al. 2002; Fairbanks et al. 2005).

^{14}C production and dilution

^{14}C is a cosmogenic nuclide (Bowman 1990). The isotope is formed from the interaction of cosmic ray neutrons with nitrogen (^{14}N) in the upper troposphere and lower stratosphere (Taylor 1987; Bowman 1990; Geyh and Schleicher 1990; Tuniz et al. 1998;

Fairbanks et al. 2005). ^{14}N reacts with neutrons to create ^{14}C and a proton (Taylor 1987). After the ^{14}C is formed it rapidly oxidizes to $^{14}CO_2$ and disseminates into all the carbon reservoirs and organisms along with the CO_2 composed of ^{12}C and ^{13}C (Taylor 1987; Bowman 1990; Geyh and Schleicher 1990; Fairbanks et al. 2005). If the rate of production is constant, then a dynamic equilibrium is formed between formation and decay and the atmospheric concentrations of ^{14}C. However, the production rate of ^{14}C has changed over time, altering its atmospheric concentration (Stuiver and Braziunas 1989; Masarik and Beer 1999; Fairbanks et al. 2005). These shifts in production can be rapid with changes to the factor of two occurring within 100 years (Hughen et al. 2004).

Changes to the rate of ^{14}C production have been linked to three dynamic processes. The most influential of these is the magnitude of the cosmic rays, responsible for the formation of ^{14}C, which can be altered by changes in the properties of the Sun's surface and the related solar winds (Stuiver and Braziunas 1989; Masarik and Beer 1999; Hughen et al. 2004; Fairbanks et al. 2005). The second process involves alterations to the intensity of Earth's geomagnetic field, which also affects the atmospheric concentrations (Hughen et al. 2004; Fairbanks et al. 2005). Increases are observed when the field strength is weakened which allows more cosmic rays to enter the atmosphere (Beck et al. 2001; Fairbanks et al. 2005). Finally, changes in the strength of the deep ocean circulation have been influential in altering the concentrations of atmospheric ^{14}C, especially between 11,000 to 14,500 yrs BP (Fairbanks et al. 2005). Because of these temporal changes in ^{14}C production, it is necessary to calibrate radiocarbon dates to account for processes that occur outside the boundaries of the initial assumptions of ^{14}C dating, including changes in atmospheric ^{14}C over time. This will be discussed later.

Concentrations also can be altered by anthropogenic activities. Atmospheric ^{14}C to ^{12}C ratios have decreased significantly since the Industrial Revolution began ~1890 when ^{14}C-depleted CO_2 began to be released into the atmosphere through the burning of fossil fuels (Bard 1988; Bowman 1990; Levin and Hesshaimer 2000). This decrease in ^{14}C is called the 'Suess Effect'. Samples dated from this time period need to be corrected for the dilution of the ^{14}C pool.

In contrast, since the 1950s, ^{14}C has been released to the atmosphere from extensive nuclear explosions (Taylor 1987; Bard 1988; Bowman 1990; Tuniz et al. 1998). Concentrations of ^{14}C in the atmosphere were increased enough to cancel the continuing dilution from the 'Suess effect' and continued to rise, decreasing the accuracy of ^{14}C dating, making it inaccurate after 1950 and very inaccurate by the mid-1960s (Taylor 1987; Levin and Hesshaimer 2000).

Radiocarbon dating and the Marine Reservoir Effect (MRE)

One of the primary concerns in paleoclimatic studies is the MRE. A reservoir effect is the alteration of the ^{14}C ratios within a reservoir relative to the atmosphere by geophysical and geochemical processes (Bowman 1990). A marine reservoir is the difference in ^{14}C age between a sample for which carbon was obtained directly from the atmosphere and a contemporaneous sample for which carbon was obtained from the ocean (Mangerud et al. 2006). Such deviations can drastically alter the dates derived from marine samples and these dates are referred to as "apparent ages" (Taylor 1987; Geyh and Schleicher 1990).

Models have been created to correct for reservoir effects, notably the work by Stuiver and Braziunas (1993), which considers the changes in the reservoir effect over space and time. Carbon reservoir modeling by Stuiver and Braziunas (1993) produced reservoir (ΔR) values ranging from 400 years in most surface areas of the global oceans to 1700 years for the deeper regions, i.e., ~1 km below the surface. However, Stuiver and Braziunas' model considered the fluxes that can occur due to solar and geomagnetic-induced ^{14}C production change, but not the effects of climatic factors on the partitioning of the ^{14}C between the different carbon reservoirs. Subsequent studies have shown that the global average MRE for the ocean surface mixed layer (usually restricted to the top 75 m and extending to 1000 m, at most), is ~400-450 years (Bard et al. 1994; Dyke et al. 1996; Björk et al. 2003; Ascough et al. 2005). This average has been used as a 'standard correction' for all marine dates from waters in the North Atlantic surrounding Canada (Dyke et al. 1996; Barber et al. 1999). This correction is relatively reliable for studies utilizing marine samples from the surface mixed layer of the North Atlantic between 40° and 70°N. In this region the ^{14}C gradient between the atmosphere and surface waters is negligible due to the homogenizing effect of the northern transport of water through thermohaline circulation and the creation of the North Atlantic Deep Water (NADW) (Mangerud 1972; Gordon and Harknesst 1992; Bard et al. 1994; Dyke et al. 1996). However, this 400 year 'standard correction' is not suitable in all circumstances. There are many instances in which a correction of 400-450 years is not appropriate, even between 40° to 70°N within the Atlantic. Reservoir corrections can range from 365 to 1900 years (Gordon and Harknesst 1992; Björk et al. 2003) because convection rates

between the atmospheric and marine reservoirs vary between marine basins and over time (Gordon and Harknesst 1992; Ascough et al. 2005)..

In addition to the differentiation in carbon isotopes that occurs from atmospheric/marine convective processes, variations in sea ice cover can further amplify the aging effects of the MRE. Sea ice can act as a physical barrier to the dissolution of CO_2 from the atmosphere to the marine surface layer (Bard et al. 1994; Franke et al. 2008a; Franke et al. 2008b). As such, the discrepancies in carbon isotopic concentrations between the marine environment and the atmosphere during periods of extended ice cover are even greater.

The 8.2 ka Event, Preboreal Oscillation, the Younger Dryas, and the Marine Reservoir Effect

The 8.2 ka cooling event, also known as the North Atlantic cooling event, (Thomas et al. 2007; Ebbesen et al. 2008) was characterized by decreased temperatures and drier conditions, most notably during the winter months (Alley and Agustsdottir 2005). It lasted approximately 160 years between 8.21 and 8.14 cal kyrs BP (Thomas et al. 2007). This event is important to understand because the climatic impacts of large melt water releases to the ocean can have implications for future rapid climate change events.

There is no definitive explanation for the onset of the 8.2 ka cooling event (Thomas et al. 2007). It is generally accepted that it was initiated by a rapid release of melt water from the Laurentide Ice Sheet, stored in proglacial Lake Agassiz, into the North Atlantic via the Hudson Strait (Barber et al. 1999). Recently, Levac et al. (2011)

and Lewis et al. (2012) have traced the path of the melt water plume along the Eastern Canadian Margin. Models show that the density changes created by the influx of fresh water would have been sufficient to slow the thermohaline circulation resulting in a cooling in the regions surrounding the North Atlantic (Wiersma et al. 2006).

Until the work of Barber et al. (1999), studies of the 8.2 ka cooling event had employed a standard correction of 400-450 years, assuming that there was no change in the reservoir effect over space or time. This led to incompatible dates between when drainage from Lake Agassiz began, the deterioration in the thermohaline circulation, and the onset of the cooling (Clarke et al. 2004). Barber et al. (1999) partly resolved the issue through creation of more dynamic reservoir corrections by using mollusk shells with known historical collection dates for the Hudson Bay and Hudson Strait regions. Barber et al. (1999) calculated that the magnitude of the reservoir effect increased from 85 ±50 years in the Eastern Hudson Straight/Labrador Sea to 310 ±50 years in the Southeastern Hudson/James Bay. This left uncertainties about the magnitude of the MRE within other areas farther south on the Eastern Canadian Margin (Newfoundland-Scotian Shelf) where studies had obtained evidence for meltwater flowing southward via the Labrador Current from the Hudson Strait and then into the North Atlantic (e.g., Li et al. 2009; Levac et al. 2011; Lewis et al. 2012). In addition to the MRE, other factors, such as the potential presence of symbiont foraminifera, need to be considered when trying to assess the magnitude of the MRE. These factors will be discussed later.

The Younger Dryas was an intense cooling event focused around the North Atlantic region, occurring at the termination of, and as a result of, the Bølling-Allerød interstadial which began around 14,600 yrs BP (Broecker et al. 1989; Weaver et al.

2003). The warming during the Bølling-Allerød interstadial increased the retreat of the Laurentide Ice Sheet and melt water drainage through the St. Lawrence, slowing the formation of the NADW and thermohaline circulation, thus, causing a cooling which lasted from 12900 – 11600 cal yrs BP (Lowell and Kelly 2008). However, there is debate about the source of the melt water with some claiming the meltwater responsible for the Younger Dryas came from the MacKenzie Delta (Murton et al. 2010). Establishing when the melt water from the Laurentide Ice Sheet began to enter the North Atlantic and affect the NADW is important to accurately map the succession of environmental/climatic events that occurred during the transition from the Pleistocene to the Holocene. Thus, having a good chronological control is crucial to establishing a cause to effect link between meltwater drainage and climatic event.

As with the 8.2 ka cooling event and the Younger Dryas, the Preboreal Oscillation is assumed to be caused by rapid meltwater incursions from the retreating Laurentide Ice Sheet and meltwater pulse(s) originating from proglacial Lake Agassiz (Teller et al. 2002; Fisher et al. 2002). The event started approximately 11300 cal yrs BP and lasted between 150 – 250 years (Fisher et al. 2002).

Considering that the Younger Dryas and the Preboreal Oscillation were probably triggered by influences similar to the 8.2 ka cooling event, the magnitude of the reservoir age would have been affected by each of the cooling periods, as was shown for the transition from the Bølling-Allerød interstadial to the Younger Dryas (Adkins et al. 1998).

Symbiosis in the foraminifera

Some foraminifera have symbiotic relationships with algae which affect test calcification rates, thus the magnitude of the MRE in a number of ways. First, the increases in the rate of calcium carbonate production resulting from photosynthesis by the algae symbionts are associated with fractionation of carbon isotopes (Kahn and Williams 1981; Duguay 1983; Lee 2004) possibly amplifying the ageing effect of the MRE. The magnitude of the age discrepancies could further be affected by the location of the foraminifera within the water column and the relative amounts of solar radiation available to the photosynthesizing symbiont and thus the rate of photosynthesis.

During the Younger Dryas, Preboreal Oscillation, and the 8.2 ka cooling event in the region of the Eastern Canadian Margin (Newfoundland and Scotian shelves) only a few planktonic and benthic species of foraminifera are known to have been prevalent. Included in the planktonic species are the formerly known *Neogloboquadrina pachyderma (sinistral),* now commonly viewed as a different species than its right coiling counterpart, *Neogloboquadrina pachyderma* (*dextral*). The two are now commonly referred to as *Neogloboquadrina incompata* and *Neogloboquadrina pachyderma,* respectively (Darling et al. 2006). Also in the planktonic assemblage are *Turborotalia quinqueloba*, *Globigerina bulloides,* and *Globigerina uvula* (Rasmussen et al. 2002; Sen Gupta 2002). The benthic species are *Bolivina subaenariensis, Globobulimina auriculata, Nonioellina labradorica*, *Elphidium excavatum* f. *clavatum,* and species of the genus *Islandiella* (Williamson et al. 1984; Denne and Sen Gupta 1989; Alve and Bernhard 1995). Of these species, assemblages of *N. pachyderma*, *E. excavatum*, *N. labradorica*, and *Islandiella* spp. seem to be the most common across the Eastern Margin (Fillon and

Harmes 1982; Williamson et al. 1984). However, despite the predominance of *E. excavatum* throughout the Eastern Margin, sometimes in excess of 40% of the assemblage (Fillon and Harmes 1982), this species was not used as a dating medium for any of the cores used in my thesis research. There are a couple of reasons for this. First, *E. excavatum* is commonly an indicator for glacial advance and decreases in salinity (Jennings et al. 2002), while *N. pachyderma*, is used as an indicator for glacial advances and sea surface temperature (SST) cooling. The latter is preferable for isotopic records because it is not a benthic species (Lubinski et al. 2001; Torres et al. 2003). Second, the prevalence of both *N. labradorica* and *Islandiella* spp. on the Eastern Canada Margin make them preferred for dating because they have robust and thick test walls requiring fewer specimens per date (Schnitker et al. 1980; Narayan et al. 2005).

The species composition of planktonic foraminifera assemblages is influenced by temperature. On the Canadian Margin, *T. quinqueloba, G. bulloides,* and *G. uvula* are more prevalent than *N. pachyderma* at SSTs >9°C. *Neogloboquadrina pachyderma* thrives at cold polar to subpolar SSTs, its abundance increasing with decreasing temperatures, and it disappears completely from the assemblages when temperatures rise above 12°C (Rasmussen et al. 2002). It is the primary, and potentially, the only planktonic calcareous foraminifera species on the Eastern Canadian Margin during the Younger Dryas, Preboreal Oscillation, and 8.2 ka cooling event.

Neogloboquadrina pachyderma is one of the most commonly used species for ^{14}C dating (Bard et al. 1994) and is not known to harbor any symbionts (Kahn and Williams 1981; Bergami et al. 2009). *Neogloboquadrina pachyderma*, has been observed to frequent areas of high nutrient concentration and to follow closely concentrations of

phytoplankton in the water column which they ingest (Stangeew 2001; Darling et al. 2007). In contrast, symbionts are usually present in foraminifera that inhabit ecological niches in which either nutrients or oxygen are limited (Lee 2004; Hill et al. 2004). Thus, dates based on *N. pachyderma* would only have been affected by the physical amplification of the MRE and potentially dietary ingestion of old carbon through meltwater detrital matter or phytoplankton around the time period of the Younger Dryas, Preboreal Oscillation, and 8.2 ka cooling event.

The benthic assemblages in the region are seemingly more influenced by deepwater salinity and dissolved organic carbon concentrations than by temperature (Williamson et al. 1984).The exception to this is *E. excavatum* which is thought to be equally affected by both temperature and salinity (Hald and Vorren 1987). However, since the limiting factors are similar for the other prevalent benthic species, and conditions during the cooling events are conducive for *E. excavatum* populations, it can be assumed that reasonable concentrations of all the predominate benthic foraminifera species mentioned could have been present within the study area during the climatic events of interest.

Of the benthic foraminifera species *Bolivina subaenariensis*, *Globobulimina auriculata, Nonioellina labradorica, Elphidium excavatum* f. *clavatum,* and the genus *Islandiella,* only *N. labradorica* and *E. excavatum* are known to retain chloroplasts from photosynthetic algae and store them in their tests (Bernhard and Bowser 1999; Pillet et al. 2011). It is thought that the retention of the chloroplasts by foraminifera within the euphotic zone subsidizes the amount of oxygen available to the foraminifera within the hypoxic substrate it inhabits (Bernhard and Bowser 1999; Hill et al. 2004). In addition,

through the process of photosynthesis, the provision of oxygen to the foraminifera would also mean increases in calcification rate through photosynthetically fixed calcium carbonate (Kahn and Williams 1981; Donner and Wefer 1994; Hill et al. 2004; Sime et al. 2005; Hamilton et al. 2008). This is of interest, for this study, in regards to *N. labradorica*, from which some of the oceanic dates were derived. However, while the chloroplasts are probably obtained from either diatoms or dinoflagellates that have descended to the substrate, it is uncertain how they benefit *N. labradorica* since it inhabits hypoxic areas beneath the euphotic zone where photosynthesis cannot occur (Bernhard and Bowser 1999; Pillet et al. 2011). Yet, Bernhard and Bowser (1999) suggest that the chloroplasts may benefit the foraminifera through metabolic processes that occur independently of photosynthesis and potentially, fix carbon. Until there are more conclusive studies of the function of chloroplasts in *N. labradorica* it can be assumed that some amplification in carbon fixation occurs. Information on *G. auriculata* is ambiguous, but reports of colour variations in some tests from the study area suggest substantial chlorophyll levels, thus the presence of chloroplasts and possibility of symbiotic relationships (Alve and Bernhard 1995). Until there are more studies regarding the symbiotic nature of *G. auriculata*, one must assume there could be some amplification of the physical and dietary MRE by fixed carbonates. *Islandiella* species can become encrusted by dinoflagellates, which could be symbiotic (Shroba 1993), but there is no conclusive evidence of calcification enhancement derived from this interaction.

Mollusks are also quite common in the study area (Gilkinson 1986; Fisheries and Oceans Canada 2010). However, there is a lack of comprehensive studies for the majority

of the mollusks comprising the assemblage there (Gilkinson 1986). Two mollusks used for radiocarbon dating of sediments from the Canadian margin are *Nucula delphinodonta* and *Macoma calcarea.* I have found no information on symbiotic relationships for *Nucula delphinodonta*. However, Kharlamenko et al. (1995) noted limited endosymbiotic interactions between bacteria and *Macoma calcarea* with most of the nutrients being provided by feeding on diatoms.

Effects of diagenetic processes on calcium carbonate carbon isotopes

Post-depositional diagenetic processes affect foraminifera and mollusks in a similar fashion (Ragland et al. 1979; Oliver et al 1996). After death, geochemical effects alter the isotopic and elemental structures of calcium carbonate shells (Mollenhauer and Eglinton 2007). The extent and magnitude of the effects vary with species and the environment. The diagenetic processes that act upon the shell material can be divided into three categories: dissolution, overgrowth, and recrystalization (Pearson and Burgess 2008). These processes can occur simultaneously and interact with one another.

Dissolution of the calcium carbonate tests, and thus the carbon isotopes, begins upon the death of the organism. In general, the initial effects of dissolution are more extensive in planktonic species which live within the top 200 m of the water column (Bé and Tolderlund 1971; Palmer et al. 1998; Lear et al. 2000). For instance, once the planktonic foraminifer has died, it loses buoyancy and sinks (Kucera 2007). Today the pH within the 0-200 m depth ranges from 8.2 at the surface to ~ 7.8 at 200 m and continues to decrease with depth. As a result, as foraminifera tests descend, the increasing acidity begins to dissolve the calcium carbonate (Pearson and Burgress 2008). The amount of test dissolution varies with species, test structure, size, and the amount of

cytoplasm that remains within the test. Larger specimens, especially those that have undergone gametogenesis, are estimated to descend at a rapid rate, reaching the substrate within a week of death. However, smaller specimens, especially those with spine ornamentation, descend at a much slower rate, reaching the substrate only after two or more weeks within the water column. The presence of residual cytoplasm will enhance the rate of deposition (Kucera 2007). Due to the differences in deposition rate, the majority of the planktonic foraminifera tests that reach the substrate are adult with the majority of the juveniles being completely dissolved within the water column prior to deposition (Kucera 2007). The extent of test or shell dissolution can be evaluated by examination under a microscope. For foraminifera, the areas most commonly affected are the chambers that were created closest to the time of death. As the magnitude of dissolution increases, areas closer to the equatorial margin demonstrate evidence of weakening and wear (Barbieri 2001). Evidence of dissolution effects become readily apparent after ¼ of the test has been affected (Barbieri 2001).

Overgrowth is the cementation of additional $CaCO_3$ on tests after they have been deposited on the substrate (Regenberg et al. 2007). Carbonate dissolved in the water column is precipitated at a rate determined by the environment. For example, regions with high limestone or chalk content have rapid rates of precipitation and overgrowth (Pearson and Burgess 2008). Overgrowth can occur on both the outer and inner surfaces of the test depending on exposure (Boyle 1983; Regenberg et al. 2007; Pearson and Burgess 2008). Overgrowth inside tests can be extensive, resulting in most of the pore space being occupied by the precipitated inorganic carbonate (Regenberg et al. 2007).

Recrystallizations also occur either after planktonic tests have begun their decent, reached the substrate, or after benthic foraminifera have died. The foraminifera tests tend to record the elemental and isotopic conditions that are present in the surrounding environment (Taylor 1987; Tuniz 1998; Torres 2003), unless there are influences from strong vital effects, pore water, etc. (Torres 2003; Costa et al. 2006). For example, the planktonic species common to the study area, *N. pachyderma* and *G. bulloides,* live near the surface of the water (Torres et al. 2003) and have no known symbionts that would cause strong vital effects (Kahn and Williams 1981; Peeters et al. 2002; Bergami et al. 2009). As such, they calcify their shells in equilibrium with the isotopic concentrations of the water column (Torres et al. 2003). Once the organism has died, however, diagenesis begins, the test begins to undergo dissolution, and once it has been deposited onto/into the substrate, isotopic exchange will occur between the test and the surrounding sedimentary environment. The isotopes are then recrystallized to form new layers of carbonate in place of those that had been dissolved (Lear et al. 2000; Torres et al. 2003; Pearson and Burgess 2008). Recrystallization differs from overgrowth as the former reformates the pre-existing isotopic structures of the old carbonate layers, while overgrowth is the precipitation and cementation of additional carbonate to the pre-existing test structure (Regenberg et al. 2007; Pearson and Burgess 2008).

Although benthic species seem to have a higher resistance to diagenetic effects they are still affected, especially by overgrowth (Boyle 1983; Lear et al. 2000). This is important, since it is generally assumed that planktonic species contain the most faithful record of the dissolved organic carbon of the sediment surface layer and thus, are

commonly used to calculate the age of the sediment (Shieh et al. 2002; Mollenhauer and Eglinton 2007).

Although the effects of dissolution create weakening and fracture of tests (Barbieri 2001), the primary problem related to ^{14}C dating is the isotopic alteration of the test structure. Since the dissolution of the tests usually begins with the most recently calcified chambers, it is the most recent record which is most readily affected by dissolution (Barbieri 2001). Since the life times of most foraminifera species are limited to a few weeks, the overall effect of outer test dissolution upon one test is not large, but can be substantial when the alterations of multiple tests is considered (Caron et al. 1987; Erez 2003).

The dissolution of the outer layers will release isotopes housed within the calcium carbonate complex (Kucera 2007). For example, moderate to severe dissolution of the outer layers could greatly alter the ratios of ^{12}C and ^{13}C to ^{14}C, thus altering the age derived from the shells/tests. The ages of the tests are further altered by the processes of overgrowth and recrystallization upon deposition to the substrate. Carbon isotopes from the dissolution of other tests and surrounding substrate can collect in and around the test further altering the overall carbon isotopic concentrations of the test. Finally, carbon isotopes from the surrounding pore water can be recrystallized to replace the areas of the shell that had been removed by dissolution, promoting the homogenization of the isotopic and elemental structure of the test with the surrounding environment (Boyle 1983; Lear et al. 2000; Torres et al. 2003; Kucera 2007; Regenberg et al. 2007).

Removing diagenetic products from carbonate shells

The contamination of the original biogenic carbonate by diagenesis can be countered by cleaning the exterior and interior of the test to remove overgrowth and recrystallized carbonate. Protocols exist (Barker et al. 2005), however they generally include the use of rigorous physical cleaning by exposing tests to ultrasonic and chemical cleaning through oxidation and reduction (Goodfriend and Stanley 1996; Torres et al. 2003; James and Austin 2008) which may dissolve the uncontaminated part of the shell (Martin and Lea 2002; James and Austin 2008). However, depending on the level of diagenetic contamination inside the test, chemical cleaning may be unavoidable because ultrasonic cleaning methods do not always remove internal overgrowths and recrystallizations (Torres et al. 2003). The best way to avoid contamination of sample materials from extensive diagenetic effects is to recognize and avoid tests which have evidence of moderate to severe diagenetic contamination.

Assessment of diagenetic effects on mollusk shells is similar to that of foraminifera in that it is partially based on visual assessment by light and electron microscopy. In addition, mollusks are assessed by trace element analysis and x-ray diffraction (Cochran et al. 2010).

Detecting alteration of original ^{14}C concentration

Alteration of the original ^{14}C concentration of shells can be determined by dating other material in the sediment. In terrestrial settings botanical macrofossils, such as leaf fragments, wood, seeds, etc. can be dated (Reimer et al. 2002; Nakamura et al. 2007; Thompson et al. 2011). However, within the marine environment this material is rare

(Brown et al. 1989), but pollen grains are present and can be dated (Vasil`chuk 2004; Vasil`chuk et al. 2004).

Pollen dating

The use of pollen grains as a substitute for macrofossils in radiocarbon dating comparisons is desirable because of the greater diversity of locations and sediment types in which pollen is abundant (Regnell 1992; Björk and Wohlfarth 2002). As with macrofossils, pollen grains are of terrestrial origin and are not affected by marine reservoir effects (Regnell 1992; Björk and Wohlfarth 2002). These two aspects make pollen an ideal candidate for making marine reservoir corrections. However, there are complications.

Brown et al. (1989) noted the importance of accurate radiocarbon dates for paleoclimatic/environmental studies and the inherent issues with utilizing total organic carbon to obtain event-constraining dates. To improve the constraining of dates, they developed a pollen concentrating method that showed statistically significant improvements in date consistency and agreement with related macrofossil dates. Sediment samples were chemically processed using standard palynological protocols, skipping the acetolysis to avoid possible carbon contamination. The samples were then alternately treated with a 2-3% NaOCl solution and sieved to retain the 44-88 µm and 44-20 µm fraction. This resulted in a sample that was comprised mostly of pollen, but also included small amounts of woody fragments, epidermal tissue, algae, and other unidentified plant tissues. The inclusion of other non-pollen materials in the concentrate was later emphasised by Regnell (1992) who showed that the sieving and chemical treatment used by Brown et al. required alteration to adequately remove non-pollen

material for satisfactory dating results. Regnell (1992) underlined the need for a more regionally and environmentally flexible method of pollen concentration.

To address the problem of residual fine organics Mensing and Southon (1999) developed a simple and inexpensive method for isolating pollen grains. They extracted pollen grains from sediment samples using a mouth pipetting system under a microscope. A test of their method on sediments from Lake Moran in California, revealed good results for sediments taken below and above the Mazama Ash layer.

Objectives

The first objective is to assess the viability of the Mensing and Southon (1999) pollen dating method on sediments retrieved from the Eastern Canadian Margin and in yet untested depositional environments, tidal wetlands. Second, if viable, the dates obtained from pollen deposited in the eastern Canadian margin sediments can be used to refine the reservoir ages for the Eastern Canadian Margin (Newfoundland – Scotian Shelf). Target periods are the onset and conclusion of the 8.2 ka cooling event, the Younger Dryas and the Preboreal Oscillation.

Chapter 2: Study Area and Methodology

Study Area

For this study I dated pollen samples from multiple sources. Samples were obtained from shallow tidal wetland deposits from southeastern James Bay, QC, Malpeque Bay, PEI and Kouchibouguacis Lagoon, NB (Figure 1). These pollen dates were compared to age models based on profiles of ^{137}Cs and ^{210}Pb in those deposits. Another set of pollen dates was obtained from buried wetland deposits near the eastern coast of James Bay, QC; these were compared to dates obtained from botanical macrofossils. Details about these cores are given below.

Ocean sediment samples used in this study (Figure 1) were from six cores retrieved along the eastern Canadian margin in which radiocarbon dates were already available from shells or foraminifera. The marine samples selected for study are from the time interval surrounding the 8.2 ka cooling event, the Younger Dryas and the Preboreal Oscillation. All of these cores have been previously studied: descriptions of the sites and cores are given below.

Ocean sediment core descriptions

The effects of diagenesis for the ocean sediment cores, especially from dissolution, can be expected to be limited, and tests and shells are usually fairly well preserved in a substantial portion of the region, where depth is between 200 m and 3000 m in depth (Parks Canada 2006a; Parks Canada 2006b). While some portions of the study area are deep enough to reach the calcium carbonate lysocline at roughly 3,500 – 4,000 m (Parks Canada 2006a; Parks Canada 2006b; Kucera 2007), the current water depths for

each of the core sites do not exceed 500 m. It is highly unlikely that the water depth changed over 3,000 m during the time interval of interest.

Core MD99-2225 is a piston core taken in 1999 in Bay of Islands, Newfoundland, located at 49.00° N, 58.08°W (Figure 1) a water depth of 104 m (Levac 2003). Four samples were ^{14}C-dated using pollen and a bivalve date is also available (Levac 2003) for one of those levels (1440-1442 cm). The core stratigraphy (Figure 2) for the uppermost section (0-14.7 m) of the 37.5 m long core shows it is composed of dark grey and black clayey silts (Levac 2003).

Core 2005033B-21PC is a piston core taken from the Inner Makkovik Bank (55.13° N, 58.17° W) in 2005 from a water depth of 352 m (Figure 1) as described by Hodder (2009). The sediments are part of the early Holocene Qeovik Silt unit (Josenhans et al. 1986). Radiocarbon dating of the core was previously done by Hodder (2009). Five depth intervals were dated using unidentified shells (Hodder 2009), most likely bivalves. The core stratigraphy shows a relatively homogenous sequence of greenish grey mud with varying percentages of sand composition and evidence of stratification at depth intervals 0-220 cm, 375-600 cm, and 900-970 cm (Figure 3) as described by Hodder (2009).

Core 2005033B-22PC is a piston core taken from the Inner Makkovik Bank (55.12° N, 58.17° W) in 2005 from a water depth of 331 m (Figure 1) as described by Hodder (2009). They are also part of the early Holocene Qeovik silt unit. Two depth intervals were ^{14}C dated using unidentified shells (Hodder 2009), probably bivalves. The

core stratigraphy shows the core is composed of very homogenous greenish grey silty clay (Hodder 2009) (Figure 4).

Core 2010023-11PC is a piston core taken from the Notre Dame Channel (51.79° N, 52.012° W) in 2010 from a water depth of 498 m (Figure 1). This core spans the equivalent sequence of sediments (Dale and Haworth 1979; Josenhans and Fader 1989) as cores 21 and 22. Two radiocarbon dates were previously derived from an unidentified shell 695 cm and unidentified foraminifera tests at 300-302 cm (Lewis et al. 2012). At the depth studied here (300-302cm), the core is primarily composed of olive grey clay (Figure 5).

Core 83033-07PC is a piston core taken from the Notre Dame Channel (50.89° N, 53.30° W) in 1983 from a water depth of 457 m (Figure 1) (Levac et al. 2011; Lewis et al. 2012). This core spans the same sediment sequence as cores 21 and 22 (Dale and Haworth 1979; Josenhans and Fader 1989). Five ^{14}C dates were previously obtained from *N. pachyderma* and *N. Labradorica* (Miller 1999; Lewis et al. 2012). The core stratigraphy shows that sediments are composed primarily of olive grey clay in the interval studied for this project (Figure 6).

Core 87033-19PC is a piston core taken from the Notre Dame Channel (50.91° N, 53.26° W) in 1987 from a water depth of 453 m (Figure 1) (Levac et al. 2011; Lewis et al. 2012). This core spans the same sediment sequence as cores 21 and 22 (Dale and Haworth 1979; Josenhans and Fader 1989). Six ^{14}C dates were obtained on *N. pachyderma* and *N. labradorica* (Levac et al. 2011; Lewis et al. 2012) and one on shell fragments (Lewis et al. 2012). The stratigraphy of the core shows the lower part of the

depth interval covered in this study (877 to 562 cm) is composed of soft, grey, calcareous clay with variable sand content, while the upper part (562 to 503 cm) is composed of firm, dark grey, and calcareous clay with minor silt content starting at 562 cm (Figure 7) (Levac et al. 2011).

Wetland core descriptions

Salt marsh cores were collected from Malpeque Bay, PEI at 46.47º N, 63.28º W) and Kouchibouguacis Lagoon in Kouchibouguacis National Park, NB at 47.08º N, 64.88º W (Figure 1) as described by Chmura and Hung (2004). The peaks in ^{137}Cs are used to identify the year 1963, which is then used for estimating accretion rates (Chmura and Hung 2004). Multiple ^{210}Pb dates were also taken for this core (Chmura 2001) and show a relatively undisrupted exponential curve, meaning that the site was relatively undisturbed, the primary reason it was chosen for this study (Figures 8 and 9).

Core AMC (Figure 1) was collected from the Eastern James Bay region (52.78° N, 78.76° W) in 2006 by Pendea and Chmura (2012) who describe the dating of its sediments using ^{137}Cs and ^{210}Pb profiles. The latter indicates regular sediment accumulation until just above 25-26 cm depth, sampled for this study (Figure 10). This depth interval was selected because of the date ^{210}Pb date reported was set prior to the inclusion of nuclear testing. This allowed for a ^{14}C and ^{210}Pb comparison without enrichment by ^{14}C isotopes from radioactive fallout.

Wetland deposits

In 2007 Pendea (2011) retrieved two additional cores (W25, 52.98ºN, 78.48ºW and W55, 53.02ºN, 78.17ºW) from the Eastern James Bay region (Figure 1) as part of a

study of wetland evolution during isostatic rebound. Both cores contain marine clay at their base and wetland deposits that reflect decreasing influence of tidal flooding and surface water inputs. Pendea (2011) determined that at the times selected for this study, W25 was a sedge carr and brown moss fen and W55 was a poor fen classified or Cyperaceae-*Sphagnum* wetland (Figure 11). In each core, seven ^{14}C dates were obtained on terrestrial macrofossils (Pendea et al. 2010; Pendea 2011). Depths corresponding to three of the dated macrofossils were selected for pollen dating (Table 2).

Methods

Pollen extraction and analysis

The ^{14}C pollen dates obtained for this study are based on bisaccate pollen grains (primarily *Pinus* and *Picea*) because they are abundant in sediments and are large (Mensing and Southon 1999). The pollen grains were extracted using the mouth pipetting system created by Mensing and Southon (1999). Samples were first concentrated using sieving and chemical digestion (Appendix 1). Individual pollen grains were extracted by dispersing small volumes of the concentrated sample with distilled water in a Petri dish and then using a narrowed glass pipette with an attached rubber tube to create a sufficient vacuum with which to manually extract and separate the grains from the rest of the residue. A dissecting scope was used to assist in the visual identification of the grains being extracted (Appendix 1).

To assess the possibility that samples contained reworked pollen, residues, from selected samples (Table 4), were mounted on slides and microscopically examined at 400X magnification. The state of preservation of pollen was classified as corroded,

degraded, crumpled, torn, or well preserved, following Birks and Birks (1980). Well preserved grains are those that showed no visible signs of exine alteration, tearing, or discolouration. Corroded grains were counted if 'etching' of the ektexine was observed. Degraded grains were those with exine demonstrating loss of defining features and opaque discolouration. Any grain that was heavily folded or creased was classified as crumpled. Finally, grains partially split or broken into pieces were classified as torn. Two separate bisaccate bladders were counted as one grain. Due to the extensive extraction of pollen that had already been done for dating, the counts obtained for degradation analysis ranged from 100-150 grains. The percent of grains in each degraded category was based on this total count. Total reworked pollen for the 503-505 cm depth of core 87033-19PC had previously been published by Levac et al. (2011).

Calibration of ^{14}C dates

^{14}C dates were calibrated using Calib 6.1.1 (Stuvier and Riemer 1993) and the Marine09 data set (Riemer et al. 2009) for marine samples and the IntCal09 data set (Riemer et al. 2009) for pollen samples. Calibrated ^{14}C dates are reported in cal yrs BP within a 2σ range and corresponding 95% confidence interval.

The ^{14}C dates were calibrated for variations in atmospheric ^{14}C concentration over time and to account for the true half-life decay of 5730 (instead of the assumed 5568 year half-life originally calculated by Willard Libby (Bowman 1990)). Calibration was initially conducted using both Calib 6.1.1 (Stuvier and Riemer 1993) and OxCal 4.1 (Bronk Ramsey 2009) to ensure that different calibration programs, with the same data set, would return approximately identical data, as suggested by Bronk Ramsey (1995) and Keenan (2012). Both Calib and Oxcal ages were the same with a variance of

approximately 1-2 years, thus dates reported here were those calculated using Calib 6.1.1(Stuvier and Riemer 1993).

Due to the modern nature of tidal wetland samples,(sediments post-dating the industrialization and the nuclear bomb testing eras), calibration was done using the Calibomb program (Reimer et al. 2004) and the NH1 data set developed by Hua and Barbetti (2004). ^{14}C results and ages from the ^{210}Pb accretion rates and the ^{137}Cs peak were evaluated using the equivalent IntCal09 calendar ages for the ^{210}Pb and ^{137}Cs and the fraction modern values (F ^{14}C) for the post-industrial and bomb data.

Corrections specific to marine ^{14}C dates

Global mean and regional ΔR

Corrections to the marine dates are required due to disequilibrium of carbon isotopic concentration between the atmosphere and the marine reservoirs. Two corrections are normally applied to marine dates: the global ocean reservoir correction (approximately 400 years), representing the mean global ocean offset from the IntCal09 curve (Stuiver et al. 2005; Reimer et al. 2009) and a regional ΔR correction to account for the regional differences in carbon isotopes for the specific region (Stuiver et al. 2005; Reimer et al. 2009). For this study, there are regional corrections of between 60-140 years referenced from McNeely et al. (2006) in the Marine Reservoir Correction Database (MRECD) at Queens University Belfast (Stuiver et al. 2012). However, the sites used for the regional ΔR calculations are located at least 100 km from the study area. The local ΔR was assessed by the comparing the marine biogenic carbonate and

pollen carbon sources without the previously mentioned ΔR values to provide the ΔR correction needed for the spatial and temporal spectrum covered in this study.

Sea ice corrections

Sea ice acts as a barrier preventing rapid diffusion of CO_2 gas between the atmosphere and the ocean thus magnifying the reservoir age in the surface layers. For this study, an additional 100 to 200 year correction needed to be applied to the marine dates to account for longer sea ice duration present in the study area during the early Holocene.

Corrections were calculated by the changes in sea ice duration from present to the time periods of interest. Sea surface conditions were reconstructed for the region by Levac (2002) and Sandercombe (2011) who used dinoflagellate cyst assemblages to demonstrate that the duration of sea ice cover was approximately 9-11 months in duration, equating to an increase of 4-6 months. The magnitude of the age increase to the ^{14}C dates was calculated using the model developed by Bard et al. (1994) (Figure 12). In the case of the Newfoundland Shelf region, the sea ice cover duration was approximately 11 months per year in the early Holocene (Lewis et al. 2009; Levac et al. 2011) compared to 5-6 months per year today, which corresponds to a 200 year magnification of the ^{14}C age (Bard et al. 1994; Levac et al. 2011).

Date comparisons

The differences in calibrated dates between the pollen and marine carbonates and macrofossils were determined using the full calibrated age ranges provided by Calib 6.1.1 (Figures 14-15 and 18-26). Ages were considered similar if the ranges had overlap. If there was no overlap between the compared age ranges, then difference in age was

calculated by using the two closest ages in the range. Age comparisons from ^{14}C-dated pollen and those based on ^{137}Cs and ^{210}Pb profiles were based on the equivalent ^{14}C ages for the ^{210}Pb- and ^{137}Cs- derived calendar years or based on the fraction Modern values (F14C; Reimer et al. 2004) for post-bomb data.

Chapter 3: Results

Twenty-one pollen samples were sent to the Keck Carbon Cycle AMS Facility in Irvine, CA for ^{14}C dating. Of these 21 samples, two (MD99-2225 2160-2162 cm and MD99-2225 2220-2222 cm) yielded too little carbon to be dated. Eleven of the 19 measurable samples were from ocean sites and eight were from wetland deposits. The final 19 dates were compared to dated material from the same depositional level, carbonate from the ocean sediments and botanical macrofossils from wetland deposits, or the age of levels based upon analysis of ^{137}Cs and ^{210}Pb profiles in other wetland deposits (Tables 1-3).

Pollen ^{14}C dates versus ^{137}Cs and ^{210}Pb dates

Ages of ^{14}C -dated pollen and tidal marsh sediments dated with ^{137}Cs and ^{210}Pb profiles do not correspond. At 13.2 cm depth in the Malpeque Bay core, the pollen returned a conventional age of -1230 yr BP and could not be calibrated. This date is well in the future of the 1963 CE date (-13 yr BP) based on the ^{137}Cs peak. The pollen age range from 22.2 cm depth is 469-664 cal yr BP, which makes the pollen 482-677 yrs older than the date calculated with ^{210}Pb (Table 1).

In the Kouchibouguacis core, the conventional date of pollen from 12.5-13.5 cm depth was -195 BP, thus also in the future and could not be calibrated. The last comparison was at 21.5-22.5cm depth with a pollen age range of 516-898 cal yr BP. This places the pollen at 499-881 yrs older than the ^{210}Pb -based age of 1933 CE (17 yr BP) (Table 1).

Sediments in James Bay core AMC were dated using a model that employed both ^{210}Pb and ^{137}Cs profiles (Pendea and Chmura 2012). The sediments at 25-26 cm depth

accumulated in a low tidal marsh environment. The pollen returned a date between 801-1051 cal yr BP and the ^{210}Pb date suggests this level was deposited 91 years before the core was extracted in 2006. As such, the ^{210}Pb age of 91 years corresponds to 1915 CE (35 yr BP) making the pollen date between 766-1016 yrs older (Table 1).

Macrofossils from James Bay buried wetland deposits

Carbon-14 dates from pollen grains and those from macrofossils reported by Pendea et al. (2010) in James Bay wetland deposits also differed (Table 2). Two depth intervals were compared in core W25. At 232.4-233.4 cm depth the age ranges from 3219-3554 cal yrs BP and the age range for the macrofossil is 2750-2916 cal yrs BP, suggesting the pollen is 303 yrs older. The second interval at 276.5-277 cm depth has a pollen age range of 2997-3168 cal yrs BP and the age range for the macrofossil is 3162-3374 cal yrs BP. Here, the pollen age is between 377 yrs younger to a 46 yr overlap with the macrofossil date. Statistically, it is more probable for the pollen to be slightly younger than the macrofossil (Table 2); however, the existing overlap denotes that the two dated samples return similar dates. This is supported by Pendea et al. (2010) who noted that the lower probability age range of the 2nd σ of 3 closely consecutive dates obtained over the 266.5 to 282 cm depth interval, showed an overlap between Beta 251813 and the next sample, 4 cm below. The lower probability 2σ age range for Beta 251813 retains the logical aging with depth profile. In core W55 (34.9-36.5 cm) the pollen age range is 689-775 cal yrs BP and the macrofossil age range is 672-783 cal yrs BP. The two age ranges only vary slightly from each other and overlap extensively making the dates returned from both samples nearly identical (Table 2).

Carbonate from ocean sediments

Analyses of ocean sediments produced some pollen dates older than the carbonate dates, some substantially younger, and some showing approximately the same age (Table 3). Pollen from core MD99-2225 (Bay of Islands) at 1440-1442 cm depth returned an age range of 7668-8021 cal yrs BP, while the carbonate of an unidentified bivalve from the same level returned an age range of 9439-10009 cal yr BP, giving a reservoir age (ΔR) of at least 1418 yrs. Core 2005033B-21PC (Inner Makkovik Bank) returned pollen dates older than the dates on the carbonate samples (Table 3). At 755-756 cm depth, the pollen age range is 8648-9252 cal yrs BP, while the age range of carbonate from the unidentified shell is 7257-7500 cal yr BP, which creates a negative reservoir age and, thus corresponds to an age reversal. At 957 cm depth the pollen age range is 9775-10558 cal yrs BP and the biogenic carbonate age range is 7422-7614 cal yr BP (Table 3). As with the 755-756 cm depth interval, there is an age reversal and a positive reservoir age cannot be obtained through the comparison. Finally, at 1038 cm depth, the pollen age range is 9483-10152 cal yr BP and the biogenic carbonate age range is 7431-7630 cal yrs BP, indicating an age reversal in the comparison and a ΔR cannot be obtained (Table 3).

Core 2005033B-22PC, also from the Inner Makkovik Bank, has one comparison at 565-566 cm depth (Table 3). However, the comparison differs from those in core 2005033B-21PC as the calibrated age ranges of the pollen and carbonate from an unidentified shell overlap. The calibration age range for the pollen sample is 7255-7937 cal yrs BP and the age range for the carbonate is 7423-7646 cal yrs BP, resulting in comparable ages. Further to the Southeast, core 2010023-11PC from the Notre Dame Channel region, has one comparison between pollen at 298-300 cm and carbonate from

an unidentified foraminifera from 300-302cm depth (Table 3). In this comparison, the pollen age range is 12720-13561 cal yrs BP and the carbonate age range is 12834-13232 cal yrs BP. The comparison shows extensive overlap between the two samples denoting very similar results.

The two comparisons from core 83033-07PC, also from Notre Dame Channel, return pollen dates younger than the foraminifera (Table 3). The pollen age range at 65-67 cm depth is 4827-5280 cal yrs BP and the carbonate age range at 63-67 cm depth is 8425-8990 cal yrs BP resulting in a minimum reservoir age of 3145 yrs. For the second pair, the age range for pollen is 7279-7619 cal yrs BP at depth 217-218 cm and the age range for the carbonate is 9928-10293 cal yrs BP at depth 206-216 cm. The ΔR for this comparison is a minimum of 2309 yrs.

The three comparisons in the last core (87033-19PC), also from the Notre Dame Channel, return pollen dates younger, older, and similar to the dates from foraminifera (Table 3). Age ranges for pollen from 503-504 cm depth and carbonates from 503-505 cm are 9664-11072cal yrs BP and 10330-12097 cal yrs BP respectively. The dates overlap at about half way through the ranges with the lower bracket of the pollen being younger than the carbonate. However, the overlap does occur in the upper range of the 97% probability within the 2σ range. Thus, while the samples can be seen as similar to having a ΔR of 520 yrs, it is highly probable that they are similar in age. The age range of pollen from 778-779 cm depth was compared to the age ranges of benthic and planktonic foraminifera from 775-780 cm depth. The uncalibrated ages place the pollen date between the benthic and planktonic dates, but once calibrated, the pollen date becomes older than either of the carbonate dates. The marine carbonate dates also retain the same

age increase with depth after calibration. The pollen age range is 11817-12383 cal yrs BP, while the age ranges for the planktonic and benthic carbonate samples are 10718-11123 cal yrs BP and 11138-11355 cal yrs BP, respectively. This shows that the age of the sample increases from the planktonic foraminifera sample through to the pollen sample with the benthic age in the middle. Thus, there is an age reversal with the pollen dates and a ΔR value cannot be calculated from the comparison. The last pair of dates is from 874-877 cm and 875-877 cm depth, for the pollen and carbonate, respectively. Calculation of age ranges produces a pollen age range of 10521-11972 cal yrs BP and a carbonate age range of 11161-11625 cal yrs BP, resulting in an extensive overlap. Thus, the returned dates are similar.

Chapter 4: Discussion

Pollen ^{14}C dates versus ^{137}Cs and ^{210}Pb dates

The cores from Malpeque Bay, PEI and Kouchibouguacis, NB, are from tidal wetlands and effects on the ^{14}C dates of pollen should vary from those of the oceanic setting. Since the ^{14}C dates are from more recent material, they are also affected by the impact of human activities on the atmosphere. However human perturbations of ^{14}C concentrations in the atmosphere do not explain all the differences in ages.

As expected, the two ^{14}C pollen dates samples from the 1960s show the effects of nuclear bomb testing. In the Malpeque Bay (13.2 cm) and Kouchibouguacis (12.5-13.5 cm) cores, the ^{14}C dates for the pollen samples corresponding to the ^{137}Cs peaks returned negative values reflecting saturation of ^{14}C isotopes from radioactive fallout (Taylor 1987; Bowman 1990; Tuniz et al. 1998). However, lower samples from both sites demonstrate unexpected dates.

The three ^{14}C dates from levels pre-dating nuclear tests (from 1915-1930s) are much older than expected (Table 1). A partial explanation for these results can be related to the introduction of large amounts of ^{14}C-depleted CO_2 beginning in the Industrial Era, the Suess effect. The highest magnitude of the Suess effect was in the 1950s when the $\delta^{14}C$ was decreased by -25‰ (Bard 1988), which results in an age range for Malpeque Bay (22.2 cm), Kouchibouguacis (21.5-22.5 cm) and AMC of -1-513, 315-660, and 673-774 cal yrs BP, respectively. The younger end of the range for Malpeque Bay falls outside of the correct time period. However, the probability is under 1‰ in the 2σ range. In addition, Kouchibouguacis and AMC are still too old. Furthermore, in the 1930s the $\delta^{14}C$ was -19‰, approximately 6‰ higher (Guilderson et al. 2005) and would have been

even higher in 1915. Thus all the samples would be too old even with full 2σ date ranges considered. As the Suess effect cannot explain the magnitude of age differences one must consider other factors. One of the most probable, is that sediments contained reworked pollen eroded from local sediments, as suggested by Roe et al. (2005) who studied pollen in a tidal marsh in Connecticut or pollen from coastal erosion of adjacent shorelines (McGuire and Chmura 2013). Evidence for this process is the reworked bisaccate and other pollen present in both Malpeque and Kouchibouguacis samples (Table 4) returning older ages. AMC core was not evaluated.

Pollen ^{14}C dates versus ^{14}C-dated macrofossils from James Bay

Radiocarbon dates of pollen and macrofossils compared favorably, however the two samples from core W25 had differences slightly higher than expected. Pendea (2011) demonstrated how the wetlands in the Eastern James Bay region underwent multiple changes over time and the difference between pollen-based and macrofossil-based ^{14}C dates of the older samples varies with the type of environment (Table 2).

The probable cause for the older pollen dates in the W25 pollen/macrofossil comparisons is reworked pollen. The sample (232.4-233.4 cm) was deposited during a fen stage (Pendea 2011). Although fens are removed from tidal influences, they do receive fluvial input (Shotyk 1996) which could be the source of reworked pollen (e.g., Chmura and Liu 1990). The second sample (276.5-277 cm) was more directly affected by fluvial processes and was deposited during the transition from tidal marsh to fen. Analysis of the first sample revealed substantial reworking (Table 4). However, while the second sample was most certainly affected by the tidal processes and reworking, it was probably not as strongly influenced as the first one. There is overlap in age ranges

between the pollen and macrofossil demonstrating a higher similarity than in the first sample (Table 2).

The pollen and macrofossil dates varied least where tidal and fluvial influences are minimized. The deposit at 34.9-36.5 cm depth in core W55 accumulated as a poor fen and returned expected similar dates (Table 2).

Pollen ^{14}C dates versus ^{14}C-dated marine carbonates

Many of the results from the ocean cores contradict the initial assumption that pollen dates would be younger than the carbonate dates. Pollen dates were younger, older, or the same age as the carbonate dates. Possible explanations are provided below.

All of the samples from the 200533B-21PC and the sample from depth interval 778-779 (pollen) and 775-780 cm (carbonate) from core 87033-19PC revealed age reversals or negative reservoir ages where the pollen was older than the carbonates. Results from cores 200533B-22PC, 2010023-11PC, and the first and last sample comparisons from core 87033-19PC, returned similar pollen dates, rather than younger dates. Since all of the samples were deposited well before any substantial anthropogenic alterations to CO_2 concentrations, other influences must be explored.

Differential bioturbation was a potential explanation for the older than expected pollen ages (Table 3) as the magnitude of bioturbation is correlated to the particle size, sedimentation rate, and the life habit of the organisms causing the bioturbation (Bard et al. 1987; Trauth et al. 1997; Thompson et al. 2000; Bard 2001). Pollen grains would be more easily bioturbated than the carbonate shells, due to their smaller particle size. However, the effects of bioturbation should be reduced under higher sedimentation rates

(Bard et al. 1987; Bard 2001), as Hodder (2009) found in core 2005033B-21PC. Sedimentation rates of 1.23 cm yr^{-1} between 583-779 cm or 1.57 cm yr^{-1} between 755-1038 cm were calculated from that core. In addition, substantial bioturbation requires macro-organism activity in the sediment which was not evident in any of the core descriptions. The more likely influences are reworked pollen (Table 4, Figure 13) in conjunction with extended sea ice cover duration during the time of sample deposition. The latter would have delayed the deposition of pollen grains to the sediment and introduced pollen grains older than the sediments into which they were deposited, further enhancing the aging effect on the pollen ^{14}C dates. However, reworked pollen is probably the most influential factor.

The influence of reworked pollen on ^{14}C dates returned by the pollen grains is especially apparent when comparing the results of core 21 and 22. Despite both cores returning unexpected dates, core 22 had a lower proportion of reworked pollen (Table 4), thus the overlap in ^{14}C dates in core 22 and the older pollen age in core 21 (Table 3). While reworked pollen can explain many of the unexpected results, not all can be completely attributed to this process.

The first sample comparison in core 87033-19PC at the 503-505 cm interval denotes an effectively similar age of the pollen and carbonate. Levac et al. (2011) correlated pollen stratigraphies within sediments of core 19 to those of Newfoundland lakes, and found that the ^{14}C age at 503 cm was in agreement with ages derived from the lake pollen zonation. For example, the age of 10268 cal BP at 503-504 cm based on carbonate and 9300 cal BP at 450 cm based on the pollen stratigraphy (Levac et al. 2011) demonstrates a reasonable age with depth curve. This would suggest that the carbonate

shells had been little affected by the MRE. However, the benthic nature of the organisms used in the carbonate ^{14}C dating would have created a heightened MRE as in MD99-2225. The 20% reworked pollen in the sample may partially explain the overlap in ages, however, the fact that the pollen age is either similar to older than the carbonate age suggests that there are other processes affecting the dating of pollen that are not fully understood at this time.

Cores MD99-2225 and 83033-07PC returned expected dates with the pollen ages younger than the carbonate ages due to the effects of MRE on the carbonates (Table 3). However, the dates from core 83033-07PC were unexpectedly younger than the carbonate dates, warranting the need to assess whether the marine or pollen date is valid. Lewis et al. (2012) and Levac et al. (2011) linked detrital carbonate peaks between cores and 83033-07PC and 87033-19PC demonstrating consistency between cores and suggesting that the carbonate dates are spatially consistent for the relevant time and similarly affected by the MRE. Since the factors of sea ice and bioturbation should be similar to the other samples in the Notre Dame Channel, another influence is responsible for the unusually large age difference. Unlike the other cores analyzed, core 83033-07PC was in warm storage for a number of years prior to being subsampled for dating. Over time, bacterial activity could have introduced modern carbon and effectively decreased the age that the pollen returned (Wohlfarth et al. 1998).

Evaluation of the MRE for the Eastern Canadian Margin

Having the most accurate ΔR values possible to account for the MRE is of vital importance to any paleoclimatic study involving the use of carbonate dating, but particularly for studies of short term, abrupt events. Each spatially and temporally defined region/depth requires a different correction. The results from this study suggest some shifts in the values that should be used in studies for specific regions and time periods, while others will require further assessment.

Using the ΔR value calculated for core MD99-2225 at 1440 cm depth, would reduce the age of the sediments at 2166 cm from 12813-11909 cal yrs BP to 11395-10491 which would shift the age of the deposits at this depth from the YD to the PBO. In relation to this study, the stark change in age interval reveals that substantial alterations in age can occur if region-specific corrections are applied. In relation to the previous study that used core MD99-2225 (Levac 2003), the corrections would also shift the ages for the depth intervals above and below the 1440 cm interval. At 710 cm, the age bracket would be shifted from 6274-5936 cal yrs BP to 4856-4518 cal yrs BP, which would place the corrected date range to the approximate time period of the 8.2 ka cooling event instead of the interstadial between the PBO and the 8.2 ka cooling event. In addition, the interval below 1440 cm would change from 12813-11909 cal yrs BP (YD) to 11395 – 10491 cal yrs BP (PBO). However, as ΔR corrections vary over time, the ΔR calculated here may not apply to both sediments above and below 1440 cm depth.

The ΔR values calculated from core 87033-07PC also suggest large magnitude shifts in original ages used in interpretations by Lewis (2008), Levac et al. (2011), and Lewis et al. (2012). Levac et al. (2011) noted the sediments at 63-67 cm were located

above a Lake Agassiz detrital carbonate bed. The ^{14}C date from those sediments was dated to circa 8.3 kya, supporting the link between the meltwater outburst from Lake Agassiz and the onset of the 8.2 ka cooling event (Barber et al. 1999). However, if the ΔR correction from this study is applied, then the ^{14}C date would be ~5 kya, and it would not be plausible to link the detrital carbonate bed and the 8.2 ka cooling event. The 206-216 cm depth interval from Core 87033-07C was used to constrain the pre-Lake Agassiz meltwater flood events. The calibrated ^{14}C age for the associated depth interval is approximately 10,000 cal yrs BP. If the ΔR correction from this study is applied, then the date for the depth interval would be approximately 7,700 cal yrs BP. Since the stratigraphic context provides strong evidence for the original interpretation by Levac et al. (2011), the new ΔR may not be appropriate. Further studies to assess the accuracy of the limited number of viable results found here are merited.

Since the results for the other cores in this study, spanning the beginning of the YD to immediately following the 8.2 ka cooling event, demonstrated either age reversals or extensive overlaps from the sample comparisons, the ΔR for those areas and time periods cannot be effectively evaluated and, thus, the ΔR values that are currently available for the region through the MRECD are the best values to utilize for studies in the region at this time. As such, past and current studies would not be affected by the findings of this study.

Chapter 5: Summary, Conclusions, and Recommendations

In this study, only 6 of the 19 pollen dates are in agreement with ^{14}C dates based on macrofossils, marine carbonates, or with the age models derived from ^{210}Pb or ^{137}Cs profiles. However, only 3 of the comparisons yielded pollen dates which were significantly younger. As in previous studies which utilized palynological samples for ^{14}C dating; many of the dates on pollen samples (either from ocean or wetland cores) are too old and were found to contain high proportions of reworked pollen grains.

However, the presence of reworked pollen grains does not translate into older pollen dates for all samples, suggesting there are processes affecting pollen dates that are still not understood. In addition, some ocean sediment cores with multiple pollen dates show age reversals that are not observed in carbonate dates, and that are not linked with changes in lithology. The near shore ocean environment is dynamic and sedimentation processes depend upon changes in the rate of water input from the coast, sea ice extent, and other factors previously discussed. As a result, near shore environments can hold considerable reworked pollen and these dynamic sedimentary environments should be avoided in pollen dating studies.

The inclusion of reworked pollen grains is problematic as there are commonly no visible signs of degradation on the grains when observing them at lower magnifications used to pick grains from the sample. Since the accuracy of ^{14}C dates from pollen grains can be greatly influenced by the reworked pollen grains, future attempts to use pollen grains for ^{14}C dating should include analyses of the condition of the pollen in the samples

to screen out samples that have too high a proportion of reworked pollen prior to the concentration of the pollen.

The 3 comparisons which did yield expected results produced significant ΔR and MRE values with the potential to shift the time periods reported in relevant studies to at least a thousand years earlier. This is important as there is the potential to alter our current understanding of the onset of rapid cooling events in the study area or at least diminish the ^{14}C dating support for those events. However, additional studies into the ΔR values for the Eastern Canadian Margin are warranted as are further consideration of the stratigraphic contexts constraining the events. The inconsistency of the results found in this study demonstrates the need for refining the understanding about pollen transportation to the substrate, storage, and movement within the sediment, and, specifically, how these processes will affect the ^{14}C dates derived from this medium.

Although results from core 83033-07PC fell within predictions of the original hypothesis; the large ΔR values were at least, partly attributable to prolonged core storage, as the young pollen ^{14}C ages could be due to bacterial degradation. More work is needed to investigate such artefacts. Certainly for now, ^{14}C dating of pollen should not be performed on cores kept in warm storage.

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Table 1: Comparison of ages of ^{14}C-dated conifer pollen extracted from tidal marsh deposits to dates determined from age models based upon ^{210}Pb and ^{137}Cs profiles in the deposits. The site numbers correspond to the locations on figure 1.

Pollen Dating					**^{210}Pb / ^{137}Cs Dating**			
UCIAMS Lab #	Depth (cm)	Fraction Modern	^{14}C Age yr BP	Cal. Age Calib 2σ range	Depth (cm)	Accretion Rate cm yr^{-1}	Date yr CE	Expected $F^{14}C$
Malpeque Bay, PEI 46.47ºN, 63.28ºW; site #1								
102908[1]	13.2	1.1661 ±0.003	N/A	N/A	13.2	0.37	1963[2,3]	1.5-2.0
102890[1]	22.2	0.9356 ±0.008	530 ±70	469-664 (1.0)	22.2	0.37	1939[2,3]	0.9792 ±0.0010
Kouchibouguacis Lagoon, NB 47.08ºN, 64.28ºW; site #2								
102891[1]	12.5-13.5	1.0254 ±0.0035	N/A	N/A	12.5-13.5	0.33	1960[2,3]	1.22 ±0.01
102909[1]	21.5-22.5	0.9160 ±0.0106	700 ±100	516-797 (0.98) 819-821(0.0012) 870-898(0.018)	21.5-22.5	0.33	1933[2,3]	0.9811 ±0.0009
AMC core Eastern James Bay, QC 52.78ºN, 78.48ºW; site #3								
105027[1]	25-26	0.8803 ±0.0037	1025 ±35	801-811(0.01) 829-860(0.06) 904-1002(0.88) 1031-1051(0.05)	25-26	0.10–0.30	1915[4]	0.9871±0.0009

N/A: Not applicable

1- This study; 2- Chmura 2001; 3- Chmura and Hung 2004; 4- Pendea and Chmura 2012. Note: (1.0) equates to the 100% probability fraction.

Table 2: Comparison of ages of **^{14}C** -dated conifer pollen extracted from buried tidal marsh deposits from Eastern James Bay to ^{14}C-dated botanical macrofossils from the same depths. The site numbers correspond to the locations on figure 1. Note: (1.0) equates to the 100% probability fraction.

Pollen Dating				**Botanical Macrofossil Dating**			
UCIAMS Lab #	Depth (cm)	^{14}C Age yr BP	Cal. Age BP Calib 2σ range	Beta Lab #	Material dated	^{14}C Age yr BP	Cal. Age BP Calib 2σ range
Eastern James Bay: Core W25 52.98ºN, 78.48ºW; site #4							
102889[1]	232.4-233.4	3160±60	3219-3229(0.006) 3240-3485(0.97) 3525-3554(0.024)	257591[2]	*Alnus* wood fragment	2710±40	2750-2878(0.997) 2914-2916(0.003)
102907[1]	276.5-277.0	2935±25	2997-3168(0.92) 3179-3208(0.08)	251813[2]	*Carex* leaf, *Eleocharis* & *Lathyrus* seeds	3060±40	3162-3190(0.05) 3201-3374(0.95)
Eastern James Bay: Core W55 53.02ºN, 78.17ºW; site #5							
105026[1]	34.9-36.5	825±20	689-775(1.0)	251810[2]	Twig	800±40	672-783(1.0)

1- This study; 2-Pendea et al., 2010.

Table 3: Comparison of dates for pollen (conifer) and marine carbonates from ocean cores. Calibrated dates are displayed as the highest probability within a 2σ range. The site numbers correspond to the locations on figure 1. B= Beta and U= UCIAMS in lab #. Note: (1.0) equates to the 100% probability fraction.

Pollen Dating				**Marine Carbonate Dating**					
UCIAMS Lab #	Core depth (cm)	^{14}C Age yr BP	Cal age (yr BP) 2σ range	Lab #	Core depth (cm)	Material dated	^{14}C age yr BP	Cal age (yr BP) 2σ range	Sea Ice Correction (yr)
Bay of Islands: Core MD99-2225 49.00ºN, 58.08ºW; 104 m depth; site #6									
102902[1]	1440-1442	7030 ±100	7668-8021(1.0)	TO8460[2]	1440-1442	Bivalve	9120 ±90	9439-10009(1.0)	150±50
Inner Makkovik Bank: Core 2005033B-21PC 55.13ºN, 58.08ºW; 246 m depth; site #7									
102886[1]	755-756	8070 ±80	8648-8675(0.017) 8684-8687(0.001) 8694-9147(0.92) 9167-9252(0.065)	B 255951[3]	755-756	Unidentified Shell	6970 ±40	7257-7500(1.0)	100±50
102903[1]	957	9060 ±130	9775-10558(1.0)	U 74202[4]	957	*Nucula delphinodonta*[4]	7135 ±20	7420-7614(1.0)	100±50
102887[1]	1038	8680 ±120	9483-9961(0.89) 9986-10044(0.037) 10055-10152(0.072)	U 74203[4]	1038	*Macoma calcarea*[4]	7150 ±20	7431-7630(1.0)	100±50
Inner Makkovik Bank: Core 2005033B-22PC 55.12ºN, 58.08ºW; 331 m depth; site #7									
102904[1]	565-566	6700 ±190	7255-7937(1.0)	B 255954[3]	565-566	Unidentified Shell	7150 ±40	7473-7587(1.0)	100±50
Notre Dame Channel: Core 2010023-11PC 51.79ºN, 52.02ºW; 498 m depth; site #8									
102906[1]	298-300	11310 ±180	12720-13511(0.999) 13556-13561(0.001)	U 96123[4]	300-302	Unidentified Foram	11765 ±25	12834-13232(1.0)	200±50

Table 3 Cont.

UCIAMS Lab #	Core depth (cm)	^{14}C Age yr BP	Cal age (yr BP) 2σ range	Lab #	Core depth (cm)	Material dated	^{14}C age yr BP	Cal age (yr BP) 2σ range	Sea Ice Correction (yr)
Notre Dame Channel: Core 83033-07PC 50.89ºN, 53.30ºW; 457 m depth; site #9									
69917[1]	65-67	4360 ±70	4827-5077(0.85) 5105-5135(0.02) 5163-5280(0.13)	B 194865[5]	63-65	*Neogloboquadrina pachyderma*	8390 ±100	8425-8990(1.0)	200±50
69918[1]	217-218	6580 ±100	7279-7283(0.003) 7287-7619(0.997)	U 20521[5]	206-216	*N. pachyderma*	9520 ±35	9928-10293 (1.0)	200±50
Notre Dame Channel: Core 87033-19PC 50.91ºN, 53.26ºW; 457 m depth; site #9									
69919[1]	503-504	9150 ±220	9664-10871(0.97) 10948-11072(0.03)	AA21750[5]	503-505	*Nonionellina labradorica*	10300 ±300	10330-12097(1.0)	200±50
69920[1]	778-779	10280 ±60	11817-12228(0.87) 12250-12383(0.13)	U 45220[5] U 45221[5]	775-780 775-780	*N. pachyderma* *N. labradorica*	10175 ±25 10450 ±25	10718-11123(1.0)/ 11138-11355(1)	200±50
69921[1]	874-877	9760 ±220	10521-10534(0.003) 10549-11843(0.97) 11860-11972(0.03)	U 45372[6]	875-877	*N. pachyderma*	10500 ±40	11161-11438(.88) 11469-11625(.12)	200±50

1- This study; 2-Levac 2003; 3- Hodder 2009; 4- Lewis, personal communication 2012; 5 Lewis et al., 2012; 6- Levac et al., 2011.

Table 4: Percentage of reworked pollen grains. Grains that showed more than one sign of degradation were classified under just one category.

Core #	Depth (cm)	Degraded	Broken	Crumpled	Total Grains	% Reworked
2005033B-21PC	1038	0	21	2	106	22
2005033B-21PC	957	6	17	1	127	19
2005033B-21PC	755-756	1	14	3	130	14
2005033B-22PC	565-566	2	2	0	102	4
Malpeque Bay	22.2	0	28	1	104	28
Kouchibouguacis Lagoon	21.5-22.5	0	14	3	100	17
W25	232.4-233.4	0	45	0	102	44

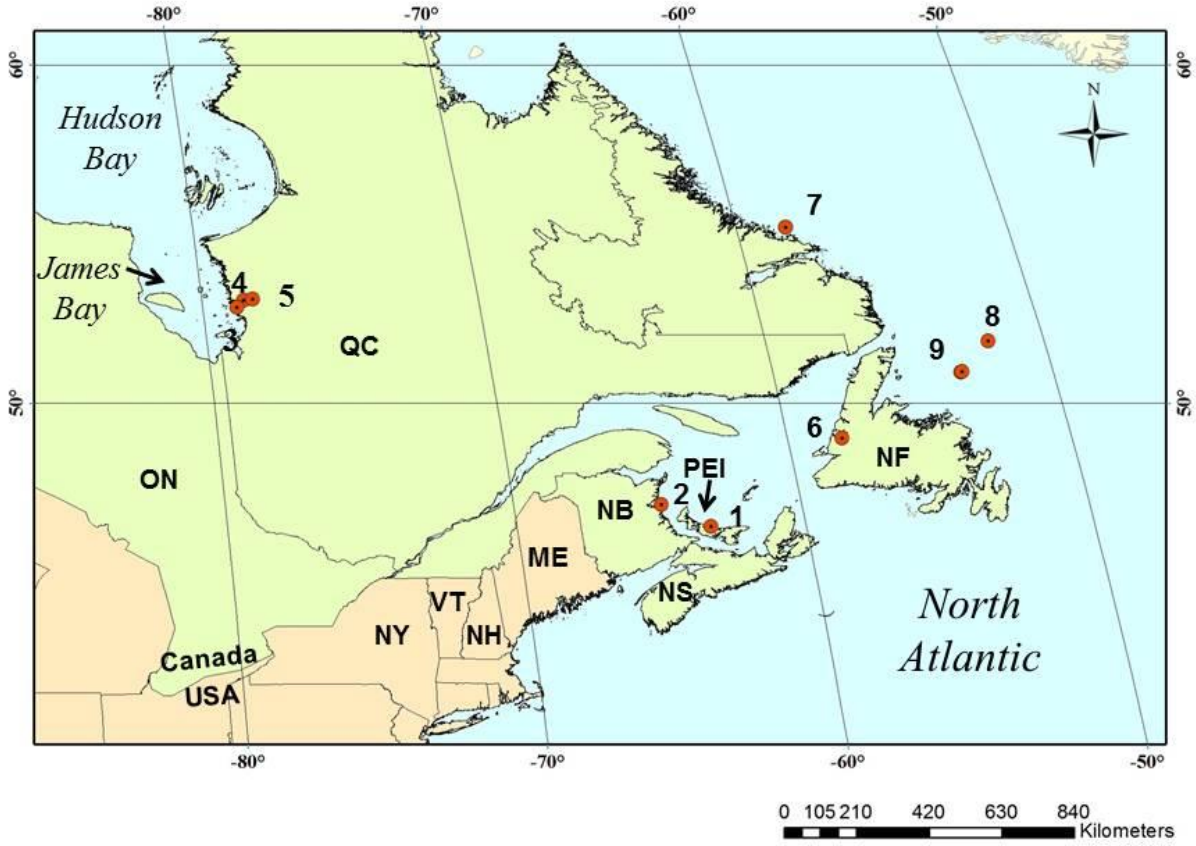

Figure 1: Location of cores used in this study: 1) Malpeque Bay, PEI; 2) Kouchibouguacis, NB; 3) Southeastern James Bay core AMC; 4) Southeastern James Bay core W25; 5) Southeastern James Bay core W55; 6) Bay of Islands core MD99-2225; 7) Inner Makkovik Bank cores 2005033B-21PC and 2005033B-22PC; 8) Notre Dame Channel core 2010023-11PC; 9) Notre Dame Channel cores 83033-07PC and 87033-19PC.

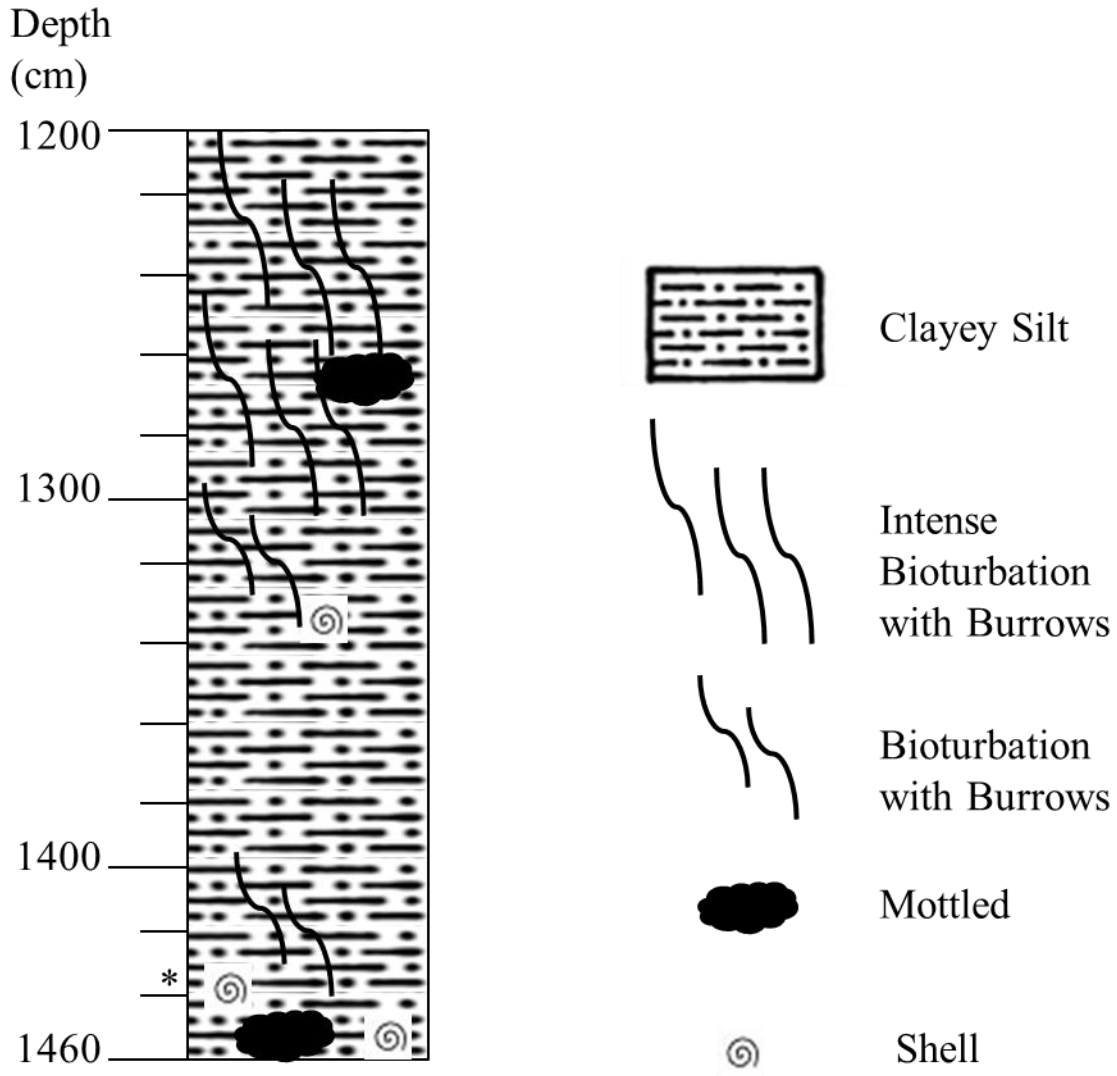

Figure 2: Core stratigraphy for MD99-2225, Bay of Islands, Newfoundland, for depth interval 1200-1460 cm. Note: * = TO8490 Bivalve

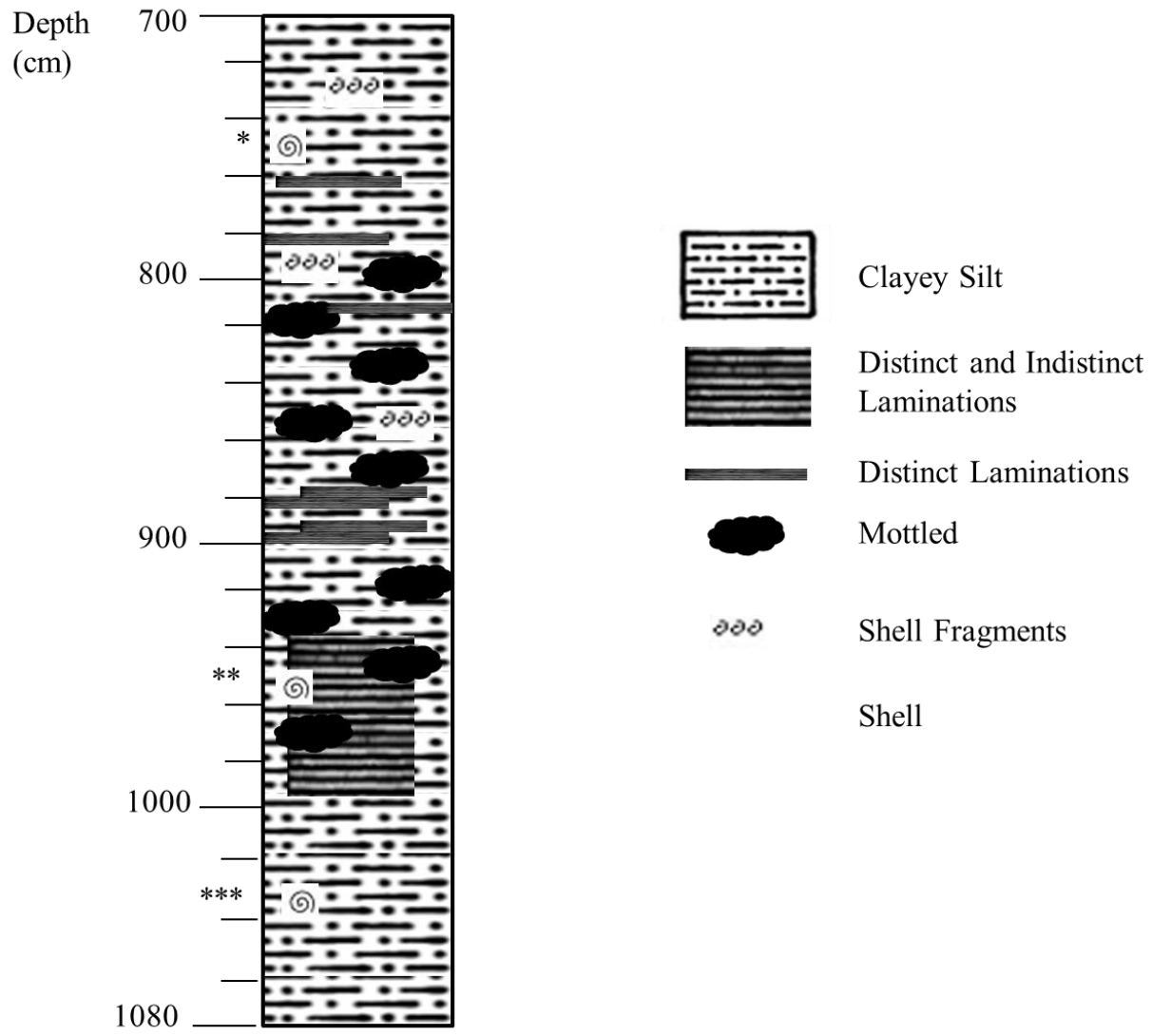

Figure 3: Core stratigraphy for 2005033B-21PC , Inner Makkovik Bank, for depth interval 700-1080 cm. Note: * = Beta 255951Unidentified Shell, ** = UCIAMS 74202 *Nucula deltinodonta*, and *** = UCIAMS 74203 *Macoma calcarea*.

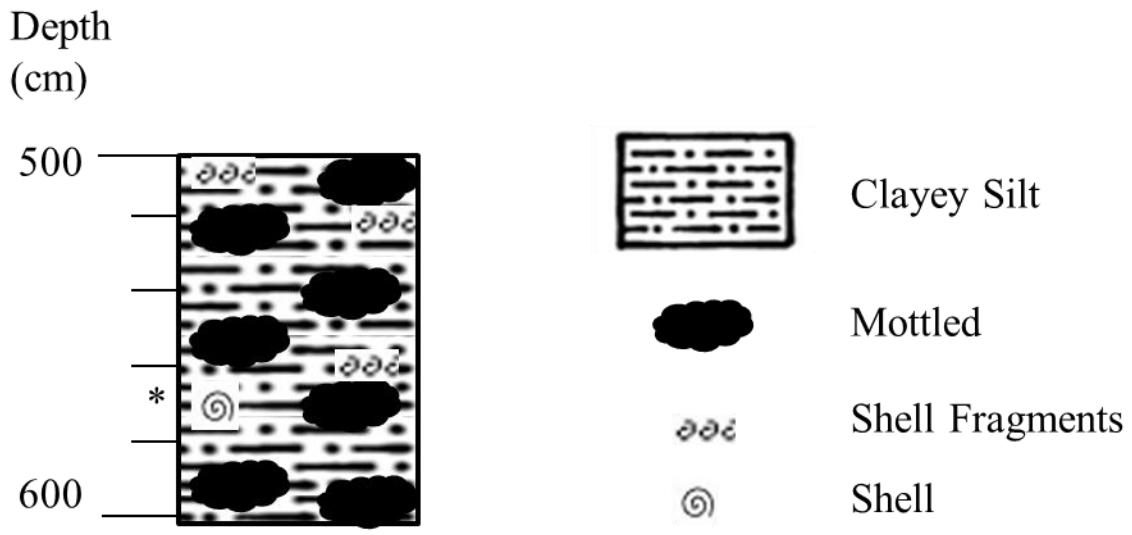

Figure 4: Core stratigraphy for 2005033B-22PC, Inner Makkovik Bank, for depth interval 500-600 cm. Note: * = Beta 255954 Unidentified Shell.

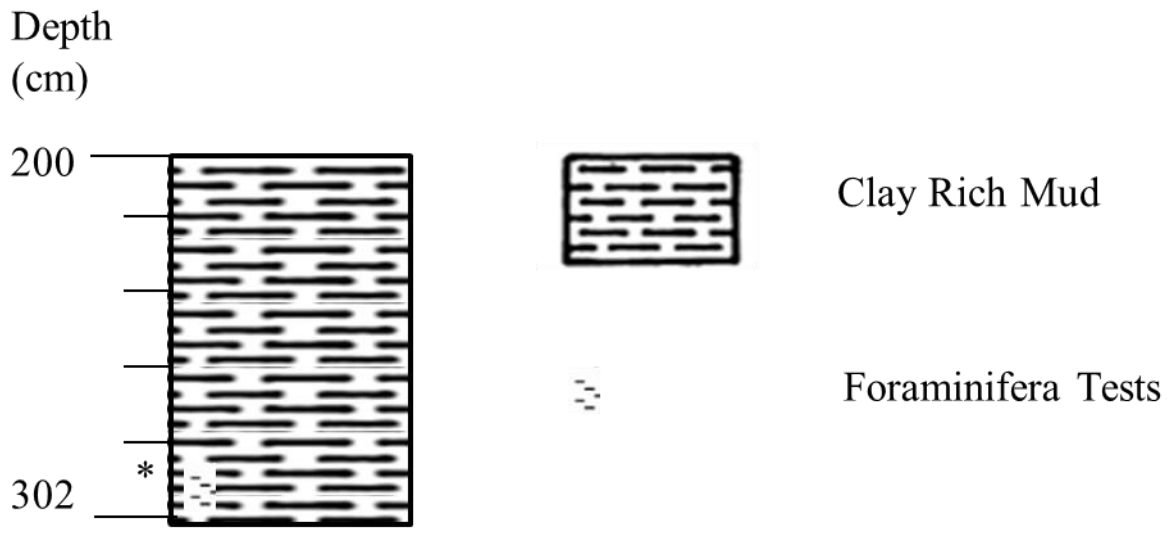

Figure 5: Core stratigraphy for 2010023-11PC, Notre Dame Channel, for depth interval 200-302 cm. Note: * = UCIAMS 96123 Unidentified Foraminifera.

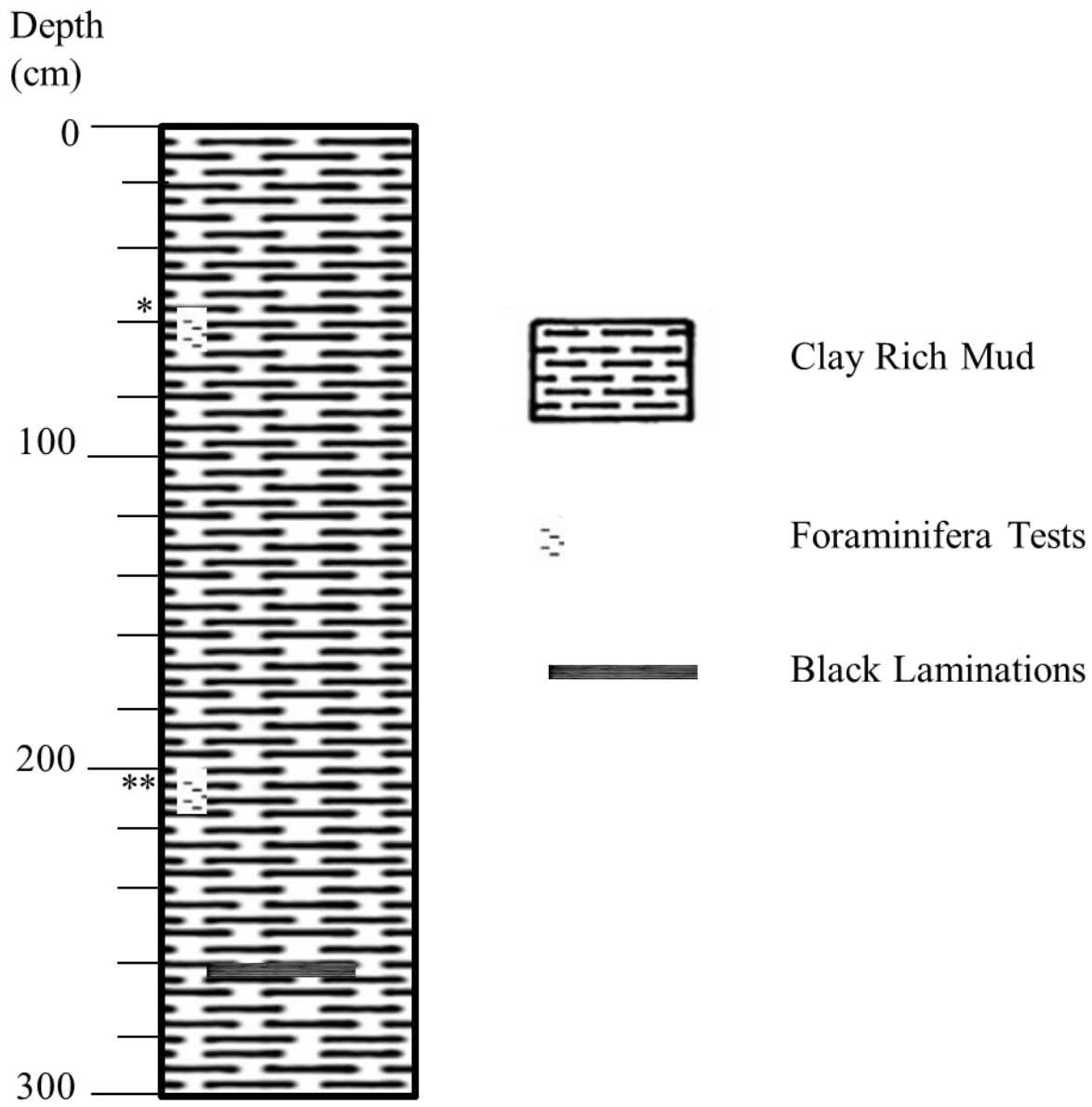

Figure 6: Core stratigraphy for 83033-07PC, NE Newfoundland Shelf, for depth interval 0-300 cm. Note: * = Beta 194865 *Neogloboquadrina pachyderma* and ** = UCIAMS 20521 *N. pachyderma*.

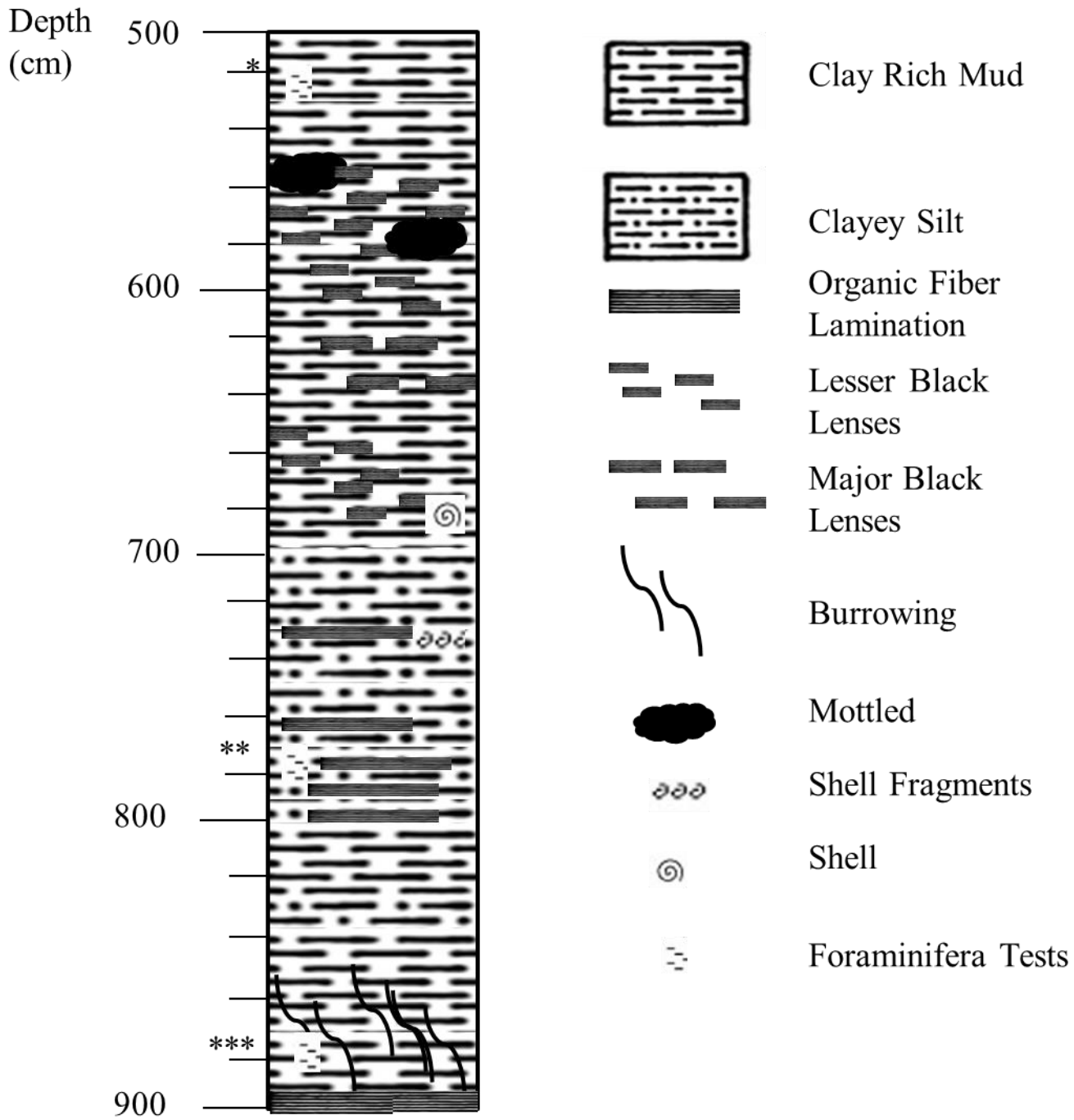

Figure 7: Core stratigraphy for 87033-19PC, Central NE Newfoundland Shelf for depth interval 500-900 cm. Note: * = AA 21750 *Nonionellina labradorica,* ** = UCIAMS 45220 *Neogloboquadrina pachyderma* and UCIAMS 452201 *N. labradorica*, and *** = UCIAMS 45370 *N. pachyderma*.

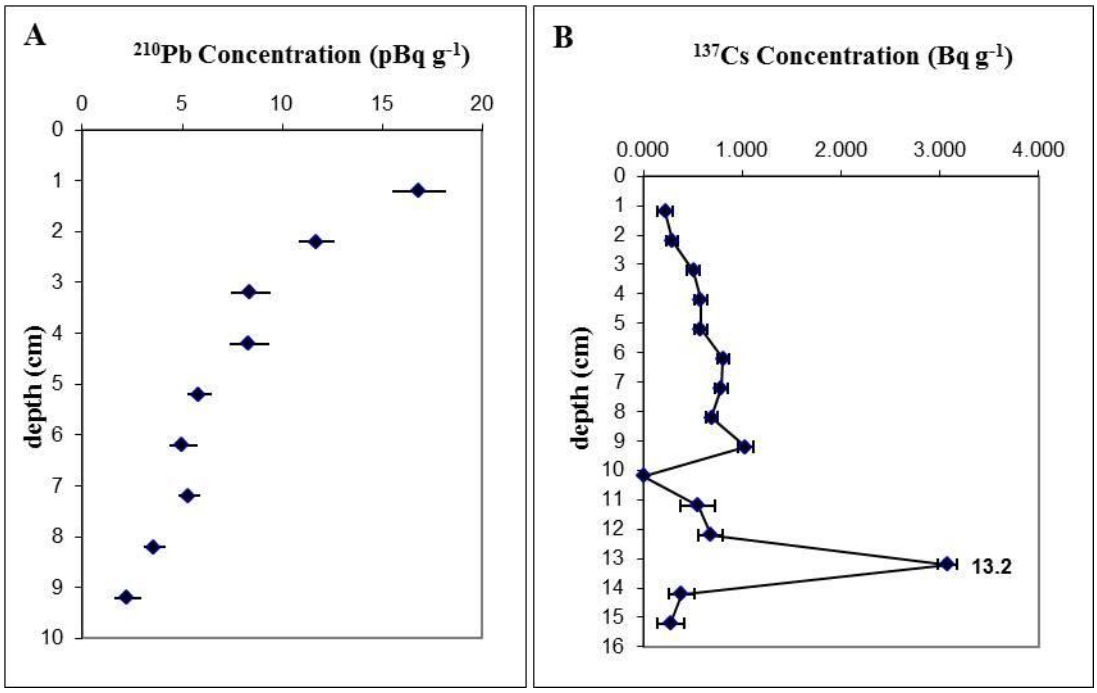

Figure 8: Radionuclide profiles for core from Malpeque Bay, PEI A) ^{210}Pb profile. B) ^{137}Cs profile. The ^{137}Cs peak at 13.2 cm corresponds to 1963 C.E. (from Chmura 2001). Error bars represent the margin of error in concentration for ^{210}Pb and ^{137}Cs.

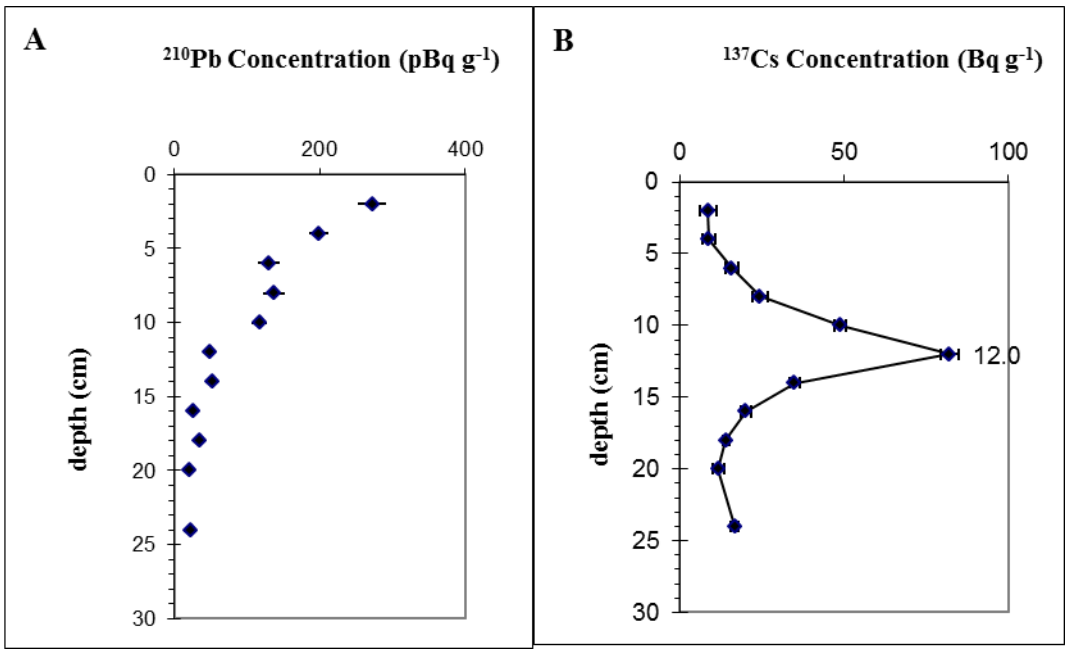

Figure 9: Radionuclide profiles for core from Kouchibouguacis, NB A) ^{210}Pb profile. B) ^{137}Cs profile for Kouchibouguacis, NB. The peak corresponds to the calendar year 1963 C.E. (from Chmura 2001). Error bars represent analytical error.

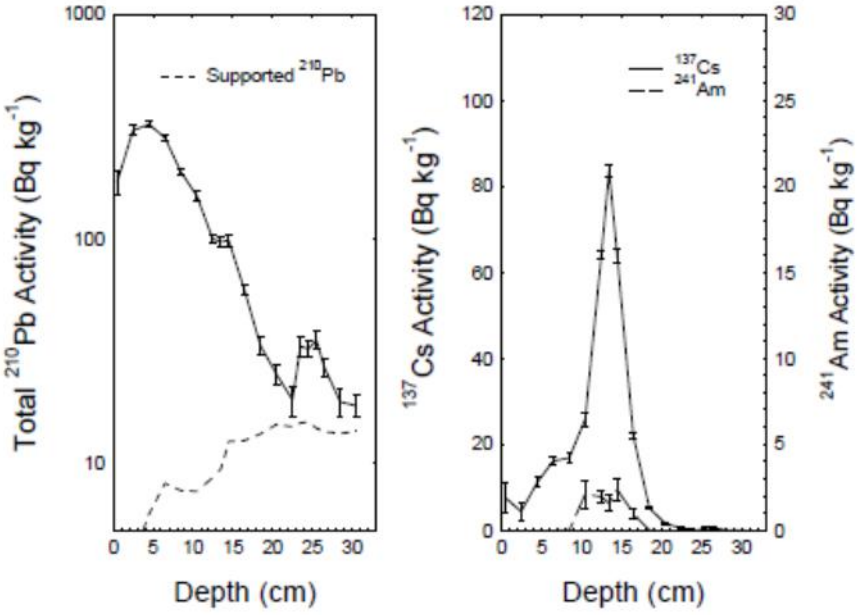

Figure 10: ^{210}Pb and ^{137}Cs activity for the AMC core. The total ^{210}Pb values denote the disturbance in the profile during the depth interval where the sample was obtained. The peak in ^{137}Cs values correspond to 1963 C.E. (from Pendea and Chmura, 2012). Error bars represent analytical error.

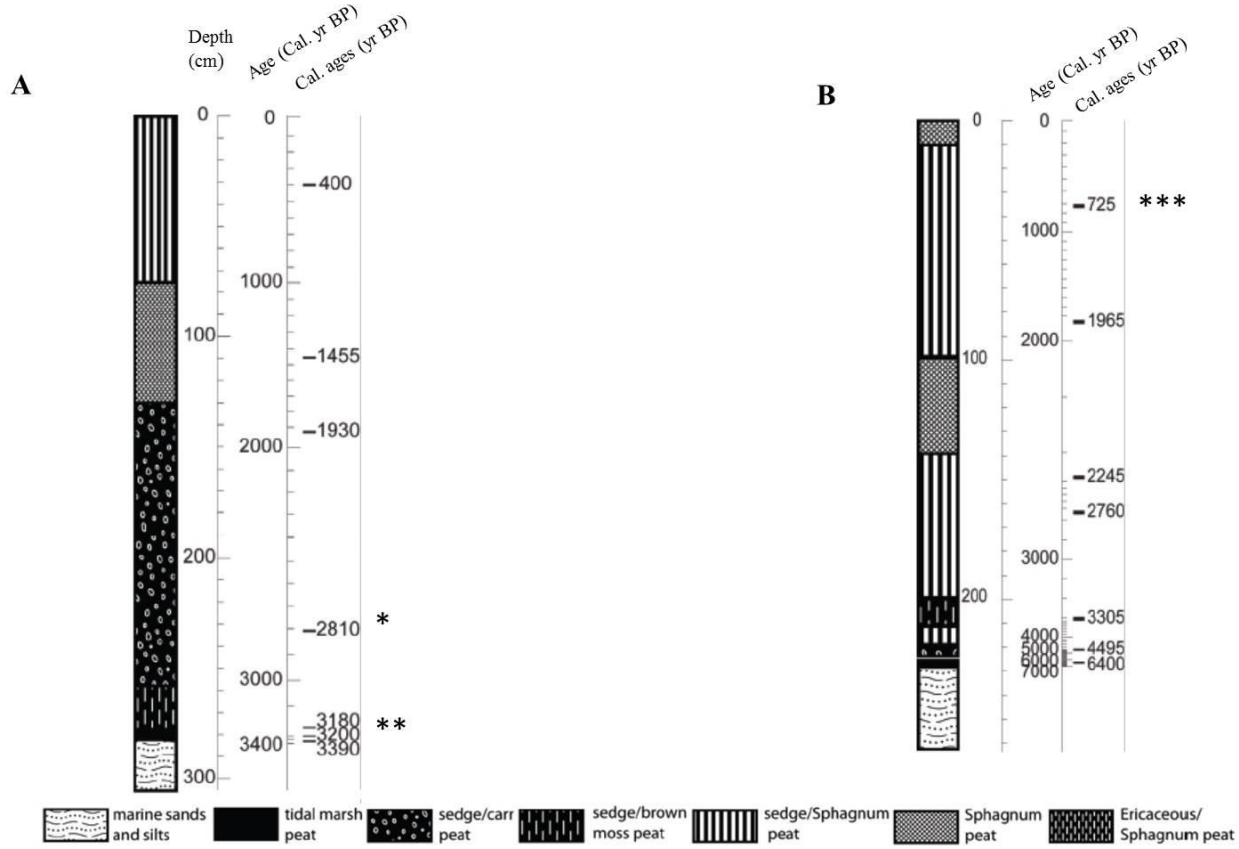

Figure 11: Stratigraphy and age vs. depth for A) core W25 and B) core W55 (Eastern James Bay region). Adapted from Pendea (2011). Note: * = the age and depth interval for Beta 257591, ** = the age and depth interval for Beta 251813, and *** = the age and depth interval for Beta 251810 reported in table 1.

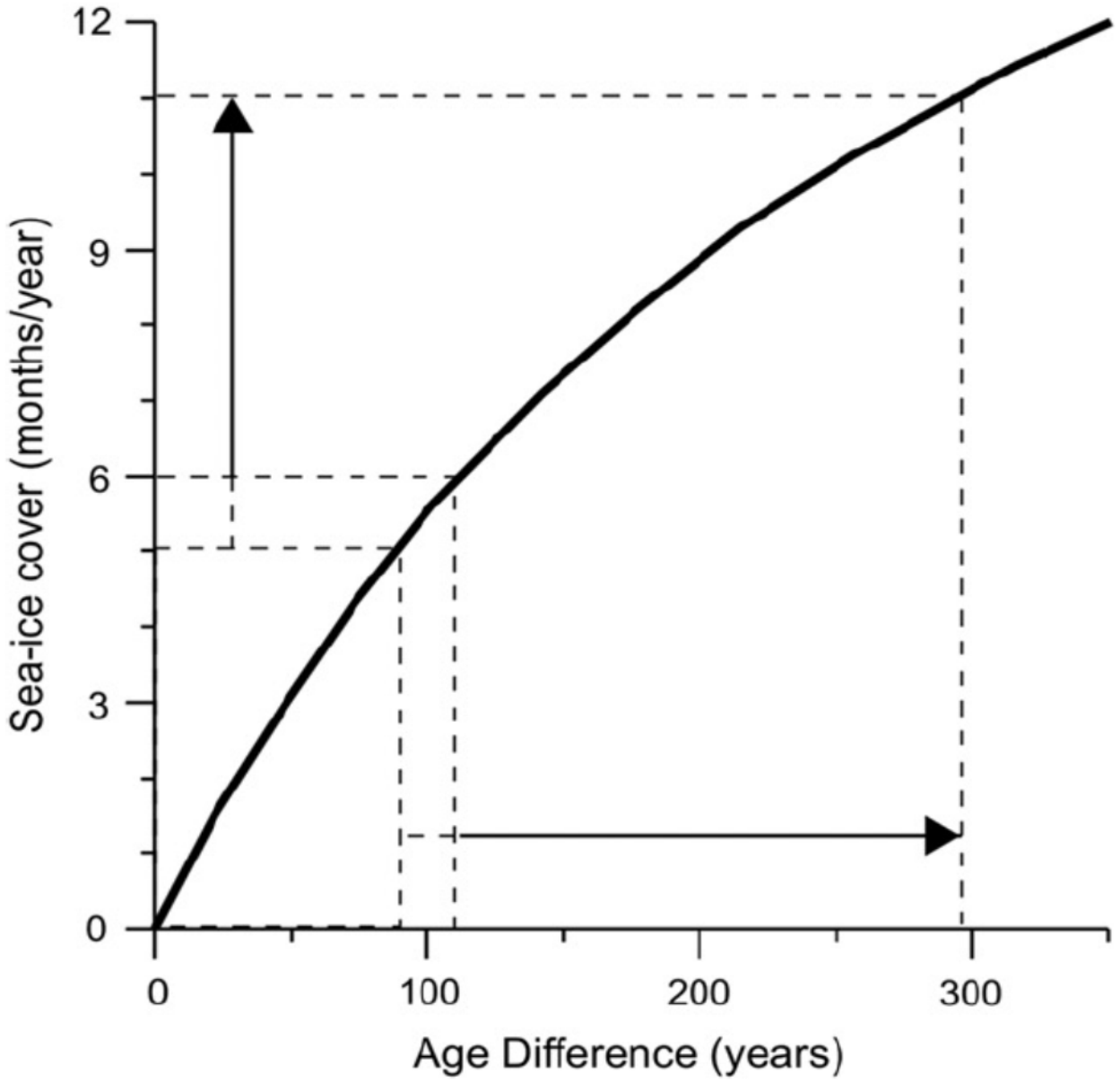

Figure 12: Relationship between extended sea ice cover duration (months beyond the present day duration) and the aging effect in surface ocean waters (figure from Lewis et al. 2012).

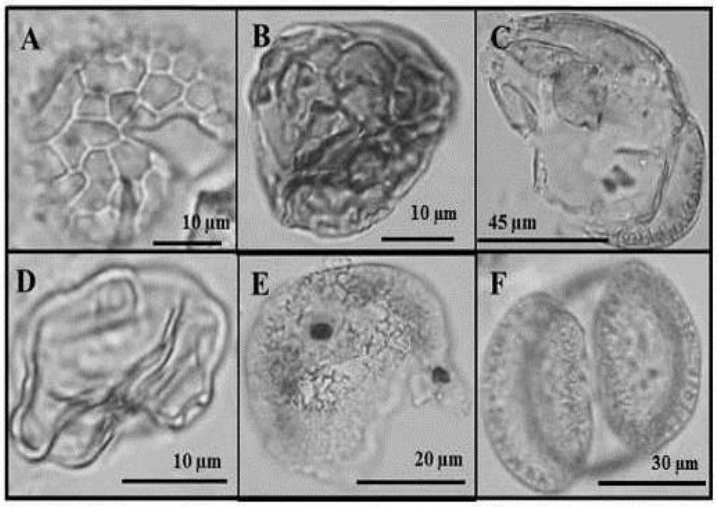

Figure 13: A) Broken *Lycopodium* spore from Core 2005033B-21PC 957 cm; splits are present at the center bottom and at the side. B) Degraded *Lycopodium* spore from Core 2005033B-22PC 565-566 cm displaying discolouration and increase in opacity. Definition loss is also present in a band just under center. C) Broken bisaccate grain from Core 2005033B-22PC 565-566 cm has lost majority of its body and a bladder. D) Crumpled *Betula* grain from Core 2005033B-21PC 957 cm is folded along center, top, and bottom left. E) Broken bladder completely separated from body. F) Well preserved *Pinus* from Core 2005033B-21PC 957 cm in which exine features are clearly defined and tears or folds are absent.

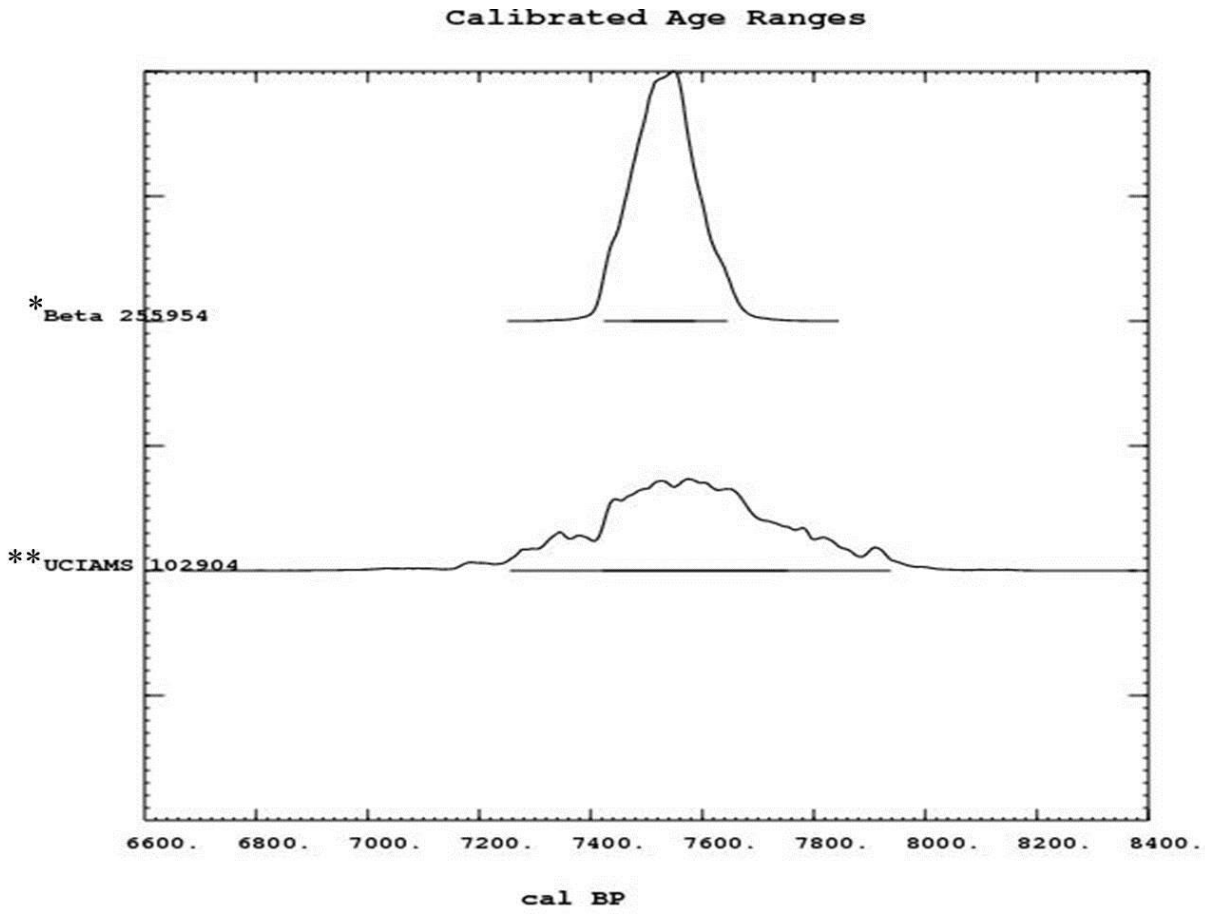

Figure 14: Comparison between calibrated age ranges for pollen and ocean carbonate samples for Inner Makkovik Bank core 2005033B-22PC, at 565-566 cm. Note: * = Unidentified Shell and ** = pollen sample.

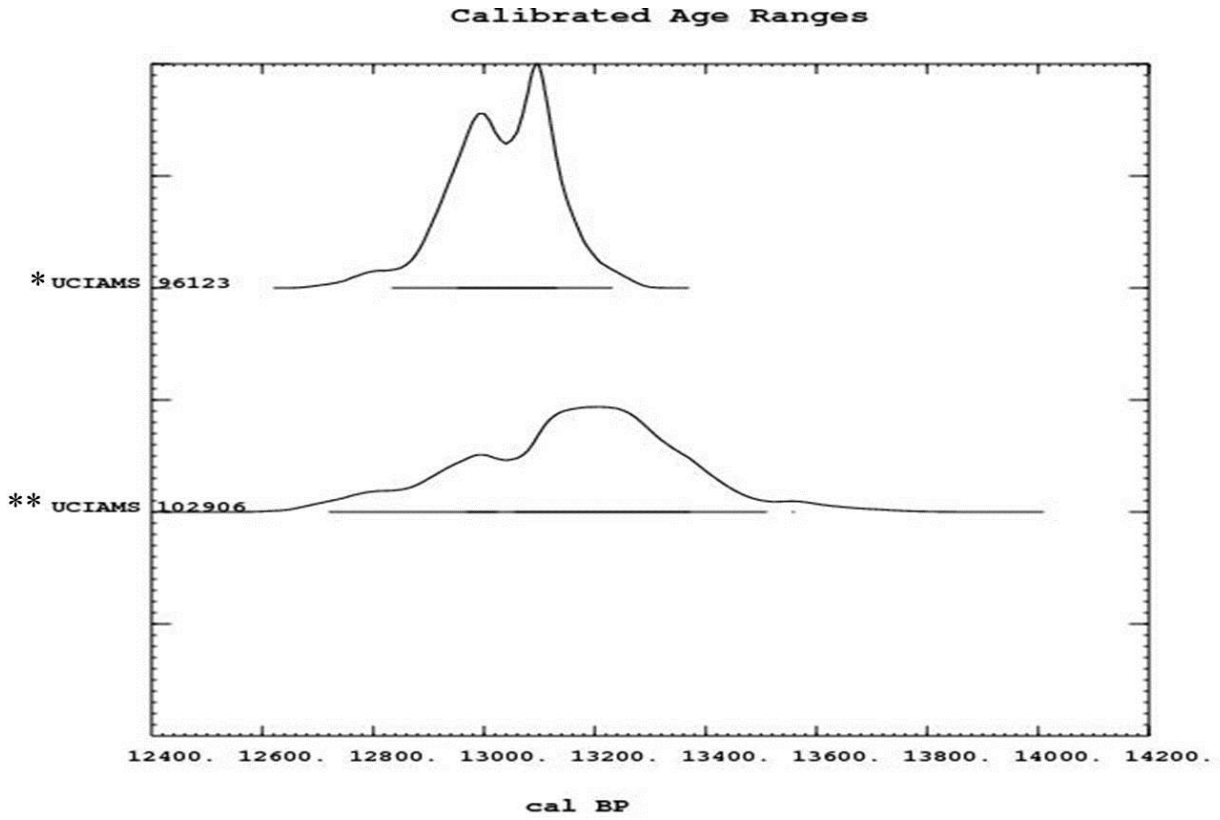

Figure 15: Comparison of calibrated age ranges for pollen (lower curve) and unidentified foram carbonate sample (upper curve) from Core 2010023-11PC. The pollen sample was taken at 298-300 cm depth and the ocean sample at 300-302 cm. Note: * = Unidentified Foraminifera and ** = pollen.

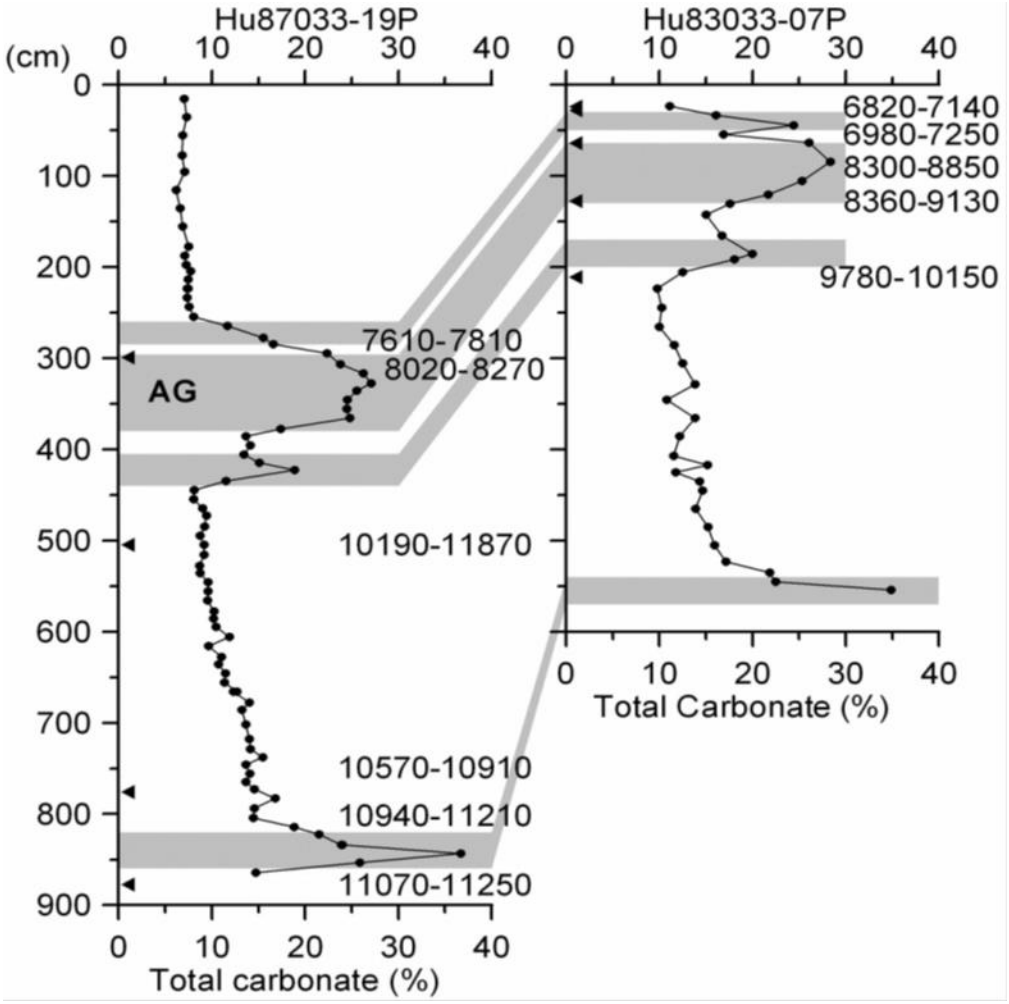

Figure 16: Total carbonate percentage in cores 87033-19PC and 83033-07PC. Grey shading highlights detrital carbonate peaks correlated between the two cores. The detrital carbonate peak corresponding to the drainage of glacial Lake Agassiz is labelled AG. Correlations between the two cores are further constrained by the calibrated ^{14}C ages (denoted by the black triangles) in each of the cores (from Levac et al. 2011).

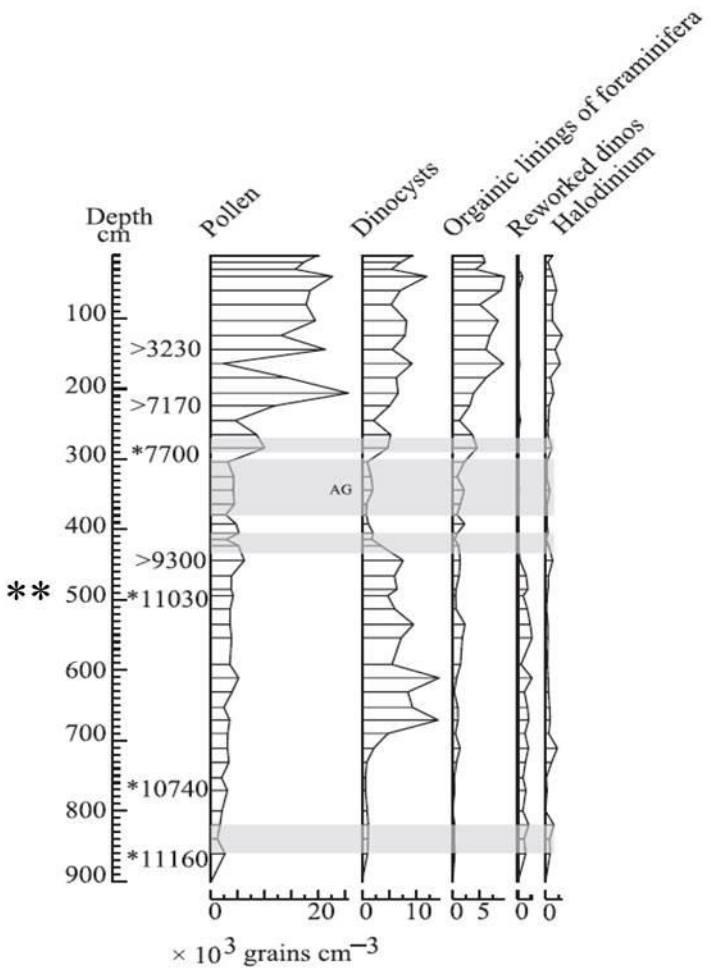

Figure 17: Palynomorph concentrations in core HU87033-19P, Notre Dame Channel. Numbers on the left are calibrated ^{14}C ages obtained from foraminifera (indicated by asterisks) or based on pollen stratigraphies. The gray shading indicates layers with high concentrations of detrital carbonate. AG corresponds to the drainage of Lake Agassiz (from Levac et al., 2011). Note: ** = UCIAMS 69919: midpoint of 10268 cal BP and a date range of 9664-11072 cal BP at depth interval 503-504 cm.

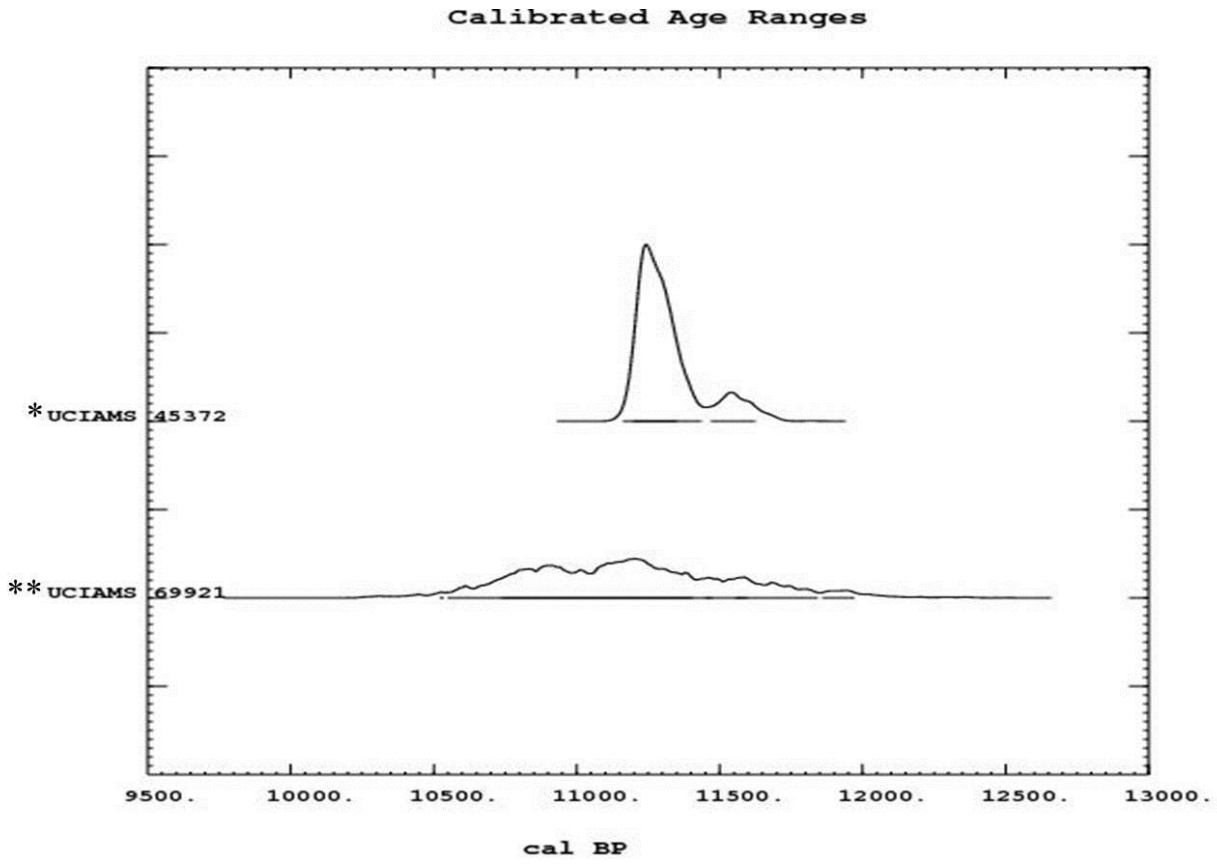

Figure 18: Comparison of calibrated age ranges of pollen and ocean carbonate samples for central Northeastern Newfoundland Shelf core 87033-19PC at 874-877 cm depth for the pollen sample and 875-877 cm for the ocean carbonate sample. Calibrated ages are labelled along the x-axis and samples labelled by lab number are located along the y-axis. Note: * = *Neogloboquadrina pachyderma* and ** = pollen.

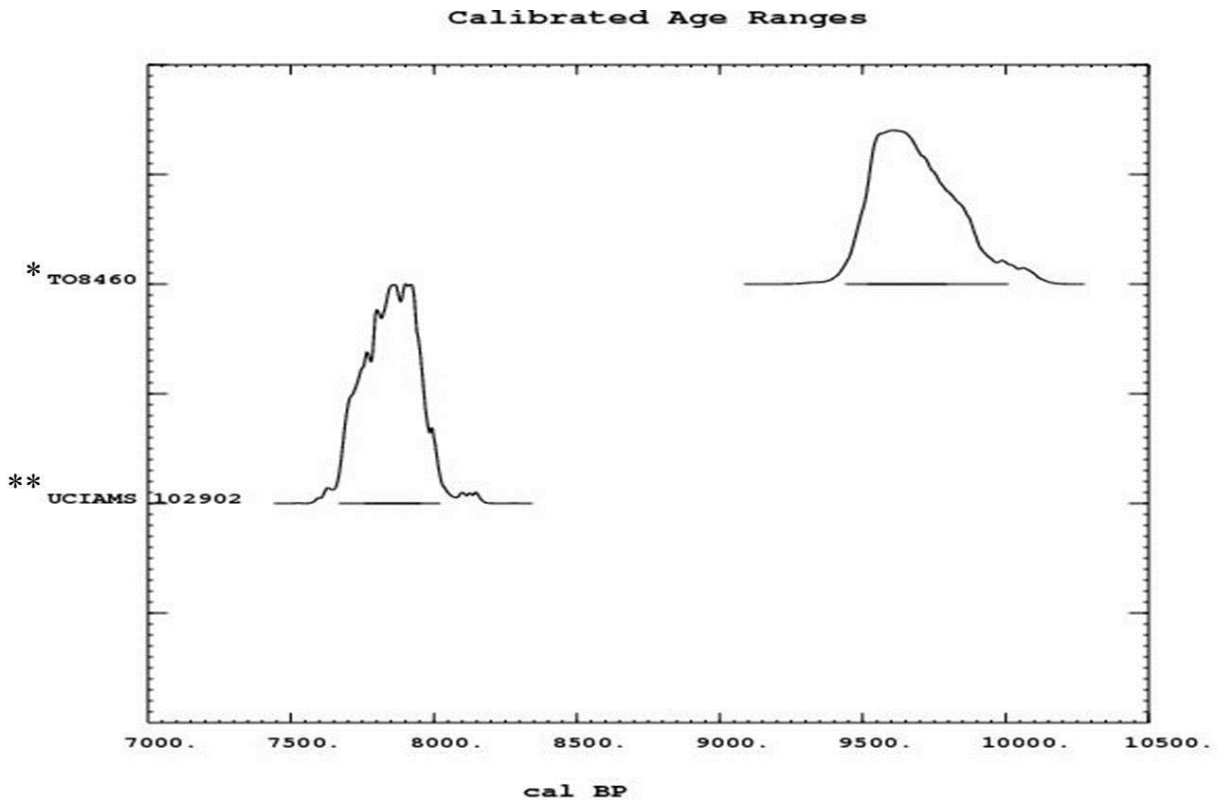

Figure 19: Comparison of calibrated age ranges of pollen and ocean carbonate samples for Bay of Islands, Newfoundland core MD99-2225 at 1440-1442 cm depth for both the pollen and ocean samples. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = bivalve and ** = pollen.

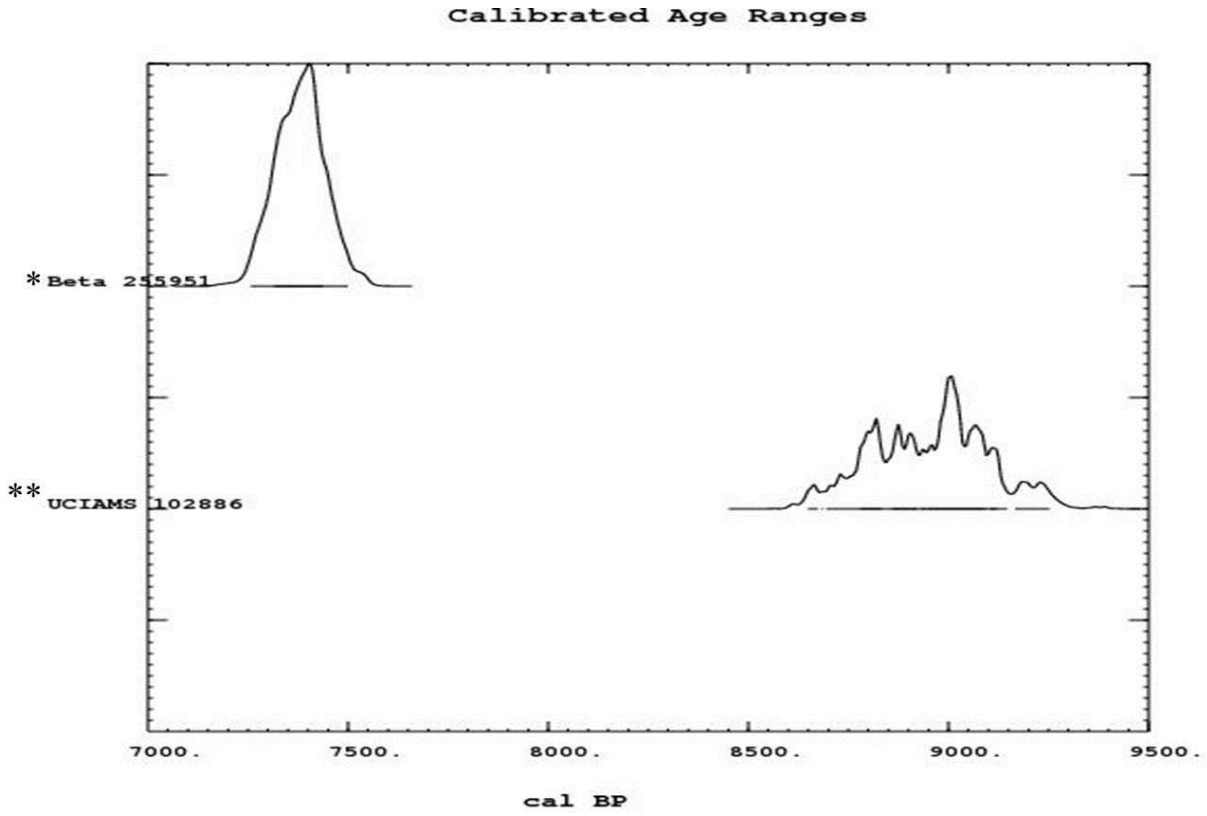

Figure 20: Comparison of calibrated age ranges of pollen and ocean carbonate samples for Inner Makkovik Bank core 2005033B-21PC at 755-756 cm depth for both the pollen and ocean carbonate samples. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = unidentified shell and ** = pollen.

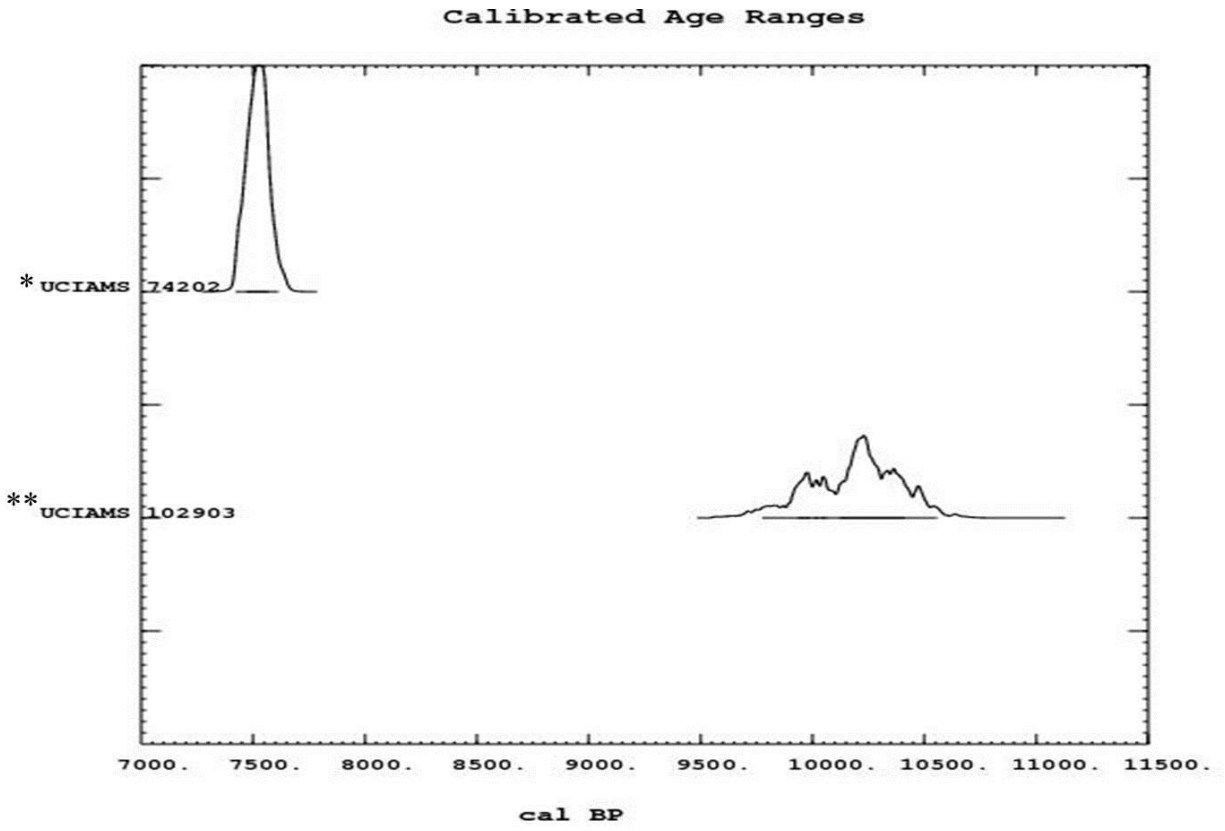

Figure 21: Comparison of calibrated age ranges of pollen and ocean carbonate samples for Inner Makkovik Bank core 2005033B-21PC at 957 cm depth. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = *Nucula delphinodonta* and ** = pollen.

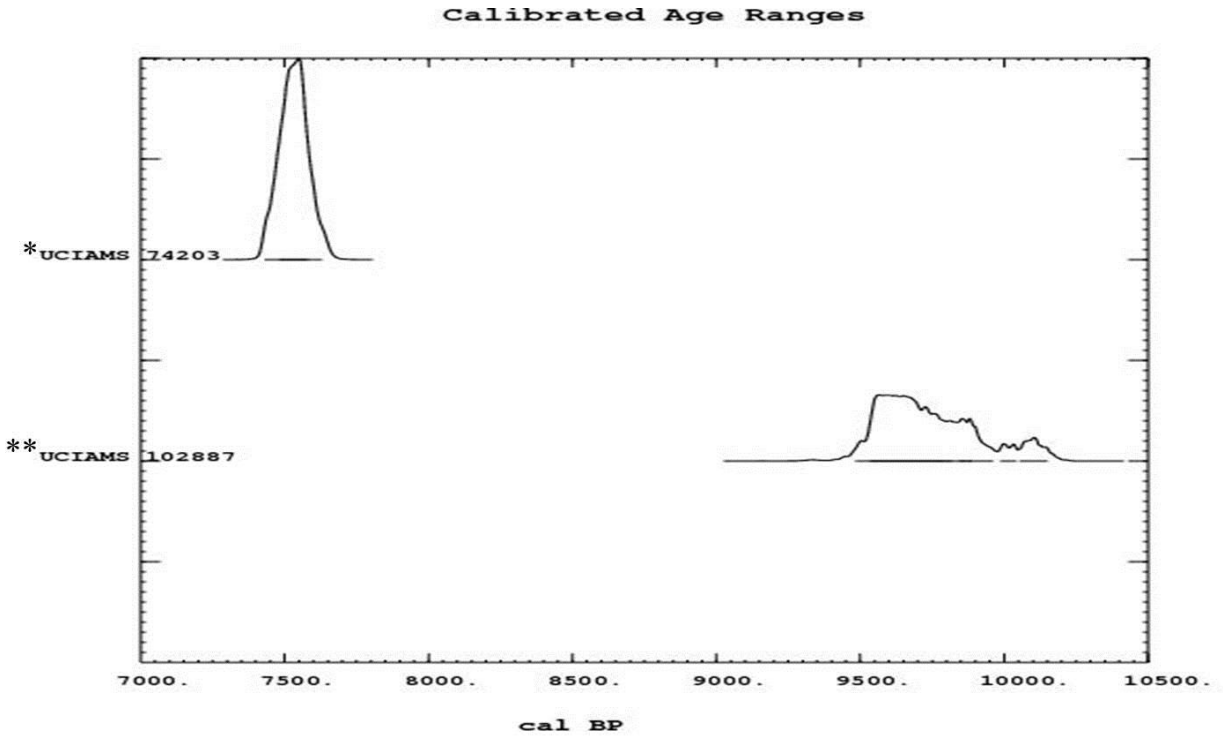

Figure 22: Comparison of calibrated age ranges of pollen and ocean carbonate samples for Inner Makkovik Bank core 2005033B-21PC at 1038 cm depth. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = *Macoma calcarea* and ** = pollen.

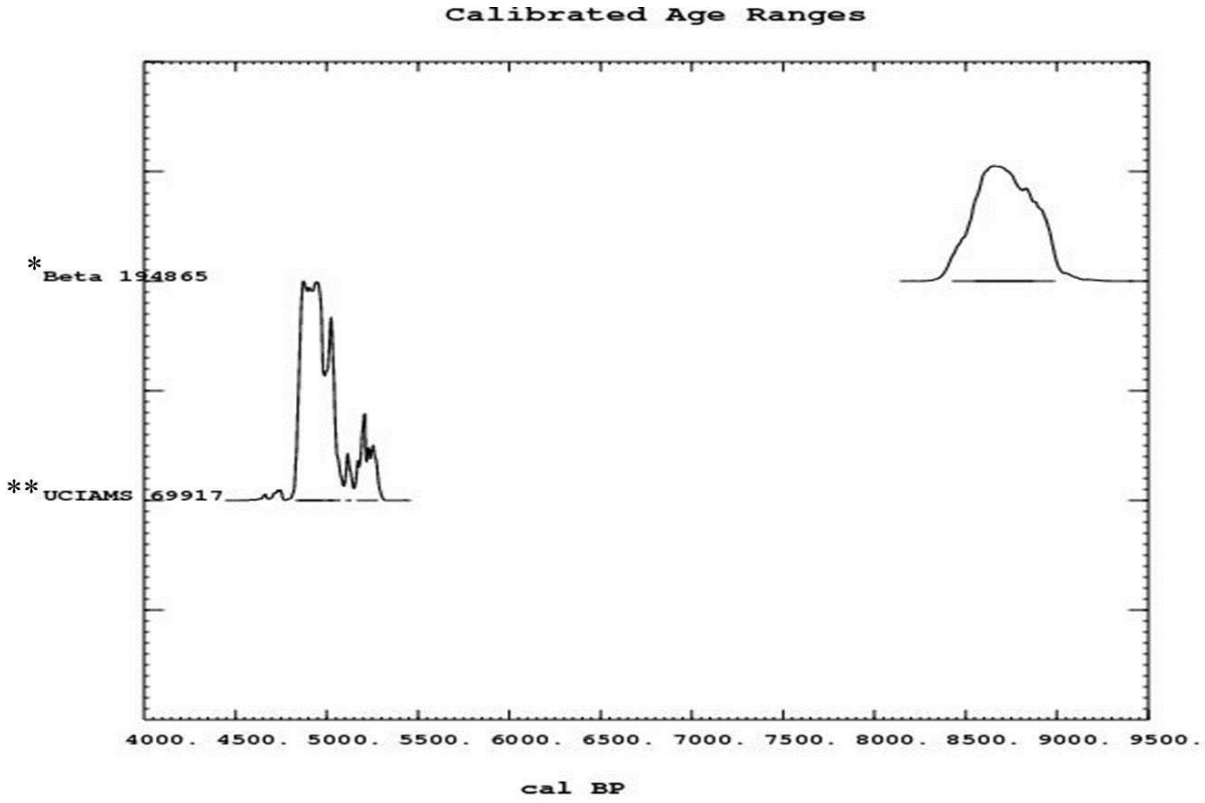

Figure 23: Comparison of calibrated age ranges of pollen and ocean carbonate samples for Northeast Newfoundland Shelf core 83033-07PC at 65-67 cm depth for the pollen and 63-67 cm for the ocean carbonate sample. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = *Neogloboquadrina pachyderma* and ** = pollen.

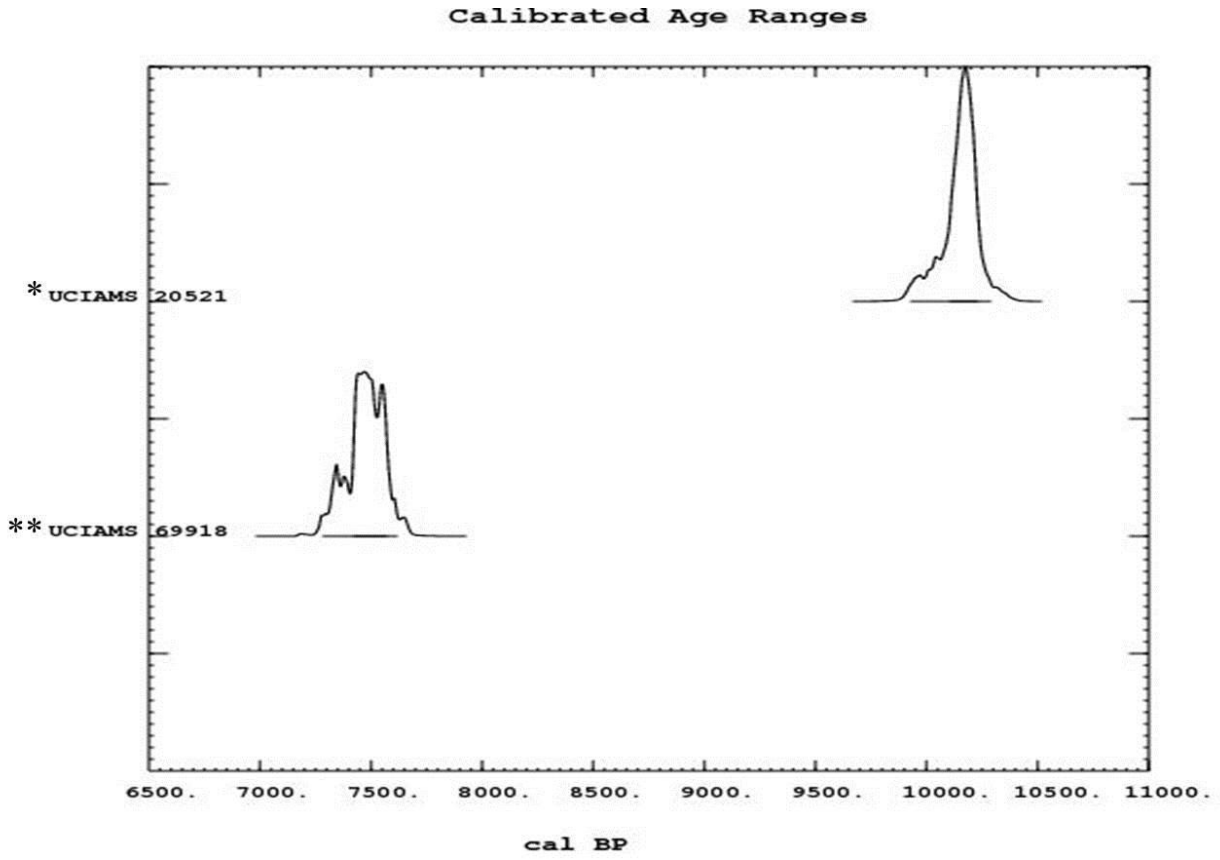

Figure 24: Comparison of calibrated age ranges of pollen and ocean carbonate samples for Northeastern Newfoundland Shelf core 83033-07PC at 217-218 cm depth for the pollen and 206-216 cm for the ocean sample. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = *Neogloboquadrina pachyderma* and ** = pollen.

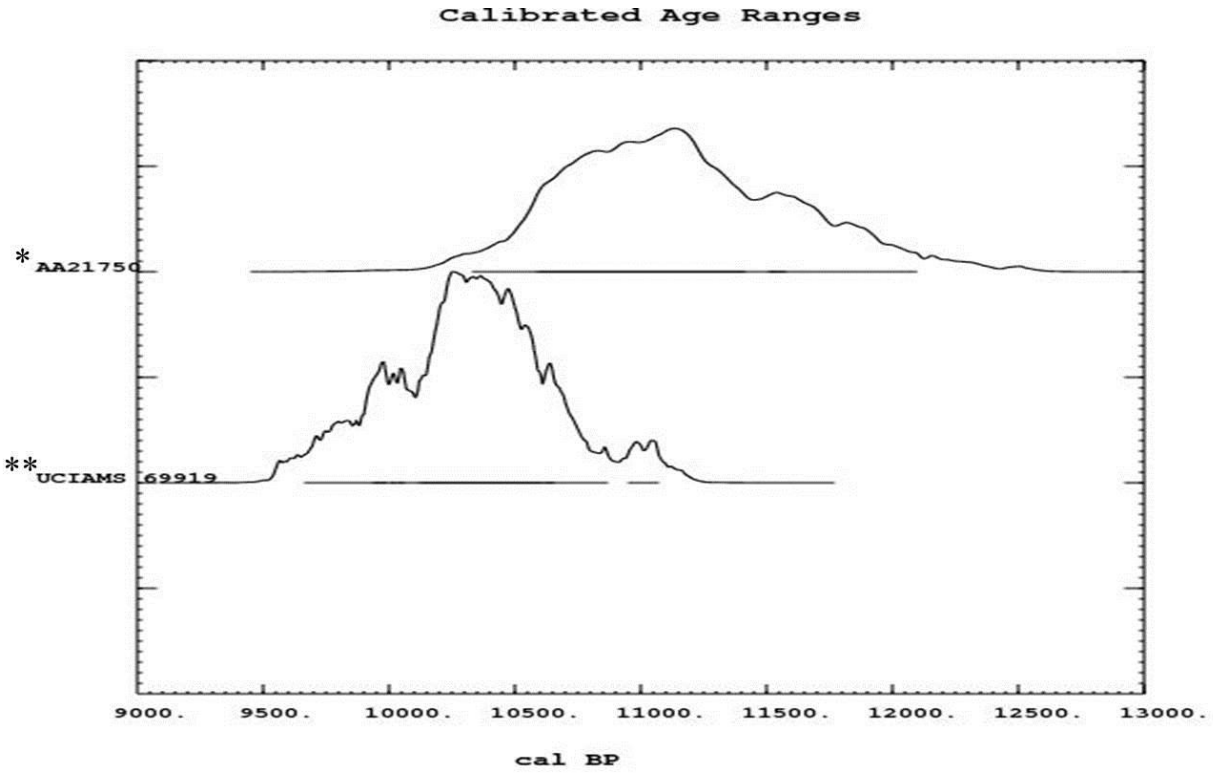

Figure 25: Comparison of calibrated age ranges of pollen and ocean carbonate samples in Central Northeastern Newfoundland Shelf core 87033-19PC at 503-504 cm depth for the pollen and 503-505 cm for the ocean carbonate sample. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = *Nonionellina labradorica* and ** = pollen.

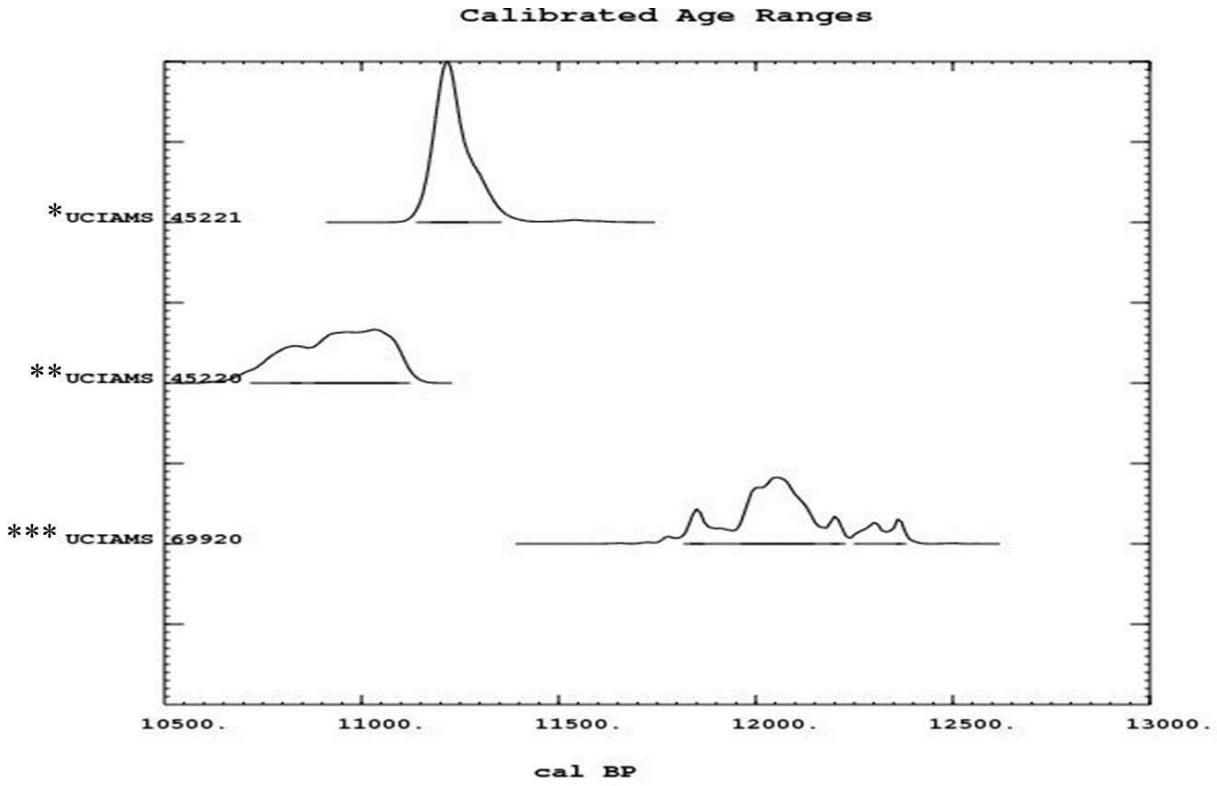

Figure 26: Comparison of calibrated age ranges of pollen and ocean carbonate samples in Central Northeastern Newfoundland Shelf core 87033-19PC at 778-779 cm depth for the pollen and 775-780 cm for the ocean carbonate samples. Calibrated ages are labelled along the x-axis and sample lab numbers are located along the y-axis. Note: * = *Nonionellina labradorica*, ** = *Neogloboquadrina pachyderma* and *** = pollen.

Appendix 1

Concentrating and Extracting Pollen

Sediment (10 cm^3) was measured by water displacement in 25 ml graduated cylinders which had been filled with 10 ml of distilled water, and weighed. A metal spatula was used to add the sediment to the cylinders. In order to guarantee that enough pollen grains were available for dating, 10 ml of marine sediment was processed instead of the standard 5ml normally used for palynological studies. The cylinders were weighed again in order to obtain the weight of the sediment being used. Next, the contents of the cylinders were emptied into 120 ml glass beakers and placed on a hot plate set on medium heat to help break the clay aggregates. The samples were kept on the hot plate for approximately 3-5 minutes and then taken off and allowed to sit for 1-2 minutes, during which time, they were constantly stirred. Once the aggregates were sufficiently broken, the samples were sieved through a 120 µm sieve using distilled water. The particles that were too large to be sieved through were emptied into a paper filter where they were allowed to dry and were then transferred into storage bags for later projects. Particles that were sieved through the 120 µm sieve were collected and then sieved over a 10 µm sieve, using a magnetic stirrer until the water ran relatively clear through the sieve. The contents of the sieve were then emptied into 50 ml centrifuge vials for chemical processing. Distilled water was used during this process to avoid contamination from the use of tap water. Depending on the lab in which the samples were processed, the protocol differed slightly. The lab of Dr. Gail Chmura at McGill University, Montreal QC, used a hot bath chemical processing protocol, while the lab of Dr. Elisabeth Levac at Bishop's University, Sherbrooke QC, used a cold standing chemical processing protocol. Both methods had similar results in pollen concentration. However,

processing times differed between the two methods. If the samples were treated with cold chemical processing, then they were processed with approximately 10 ml of 10% Hydrochloric acid (HCL) for a maximum of 30 minutes, or until reactions stopped, to remove carbonates. After being treated with HCL, the samples were centrifuged for 10 minutes at 3,500 rpm. Spent/excess HCL was decanted out of the centrifuge vials into appropriate waste containers. Samples were then rinsed twice with distilled water and decanted, with the samples being centrifuged prior to each decanting.

Following treatment with HCL, the samples were transferred into 10 ml centrifuge vials for Hydrofluoric acid (HF) treatments, unless the sample was not disseminated sufficiently. Approximately 5 ml of 49% HF solution was added and allowed to sit for approximately 12 hours. This removed the excess silicates in the samples. As with the HCL, following acid digestion, the samples were centrifuged for 10 minutes at 3,500 rpm and then the HF was decanted into appropriate waste containers. Distilled water was then added twice to the samples which were then centrifuged in between each application of distilled water and decanted in order to rinse out any remaining HF within the samples.

If hot chemical processing was used, then the samples were put into 50 ml vials, unless the sample was small enough to allow for processing in 10-15 ml vials. A hot water bath was prepared in a 500 ml Pyrex beaker placed on an electrical hotplate to bring the water to a boil.

Approximately 10 ml of HF was directly added to each of the vials that were put into the bath. After the HF had been added, the samples were mixed using a stir stick. Second, the vials were placed in the hot water bath for approximately 20 minutes during which, the samples were stirred every few minutes. New sticks were used in the event that the original sticks began to

disintegrate during the processing. Third, once the 20 minutes were up, the samples were centrifuged for 3 minutes at 3000 rpm (the differences in time and rpms was due to the make of the centrifuge used in each lab). The effectiveness on sample settling is approximately the same. Finally, unlike in cold processing, the samples were not rinsed and were immediately prepared for hot HCL treatment. The reason for this was for the HCL to remove the silicofluoride precipitated during the HF digestion.

Using the same vials as in hot HF, approximately 10 ml of 10% HCL solution was added to each of the samples that had been processed with HF. Second, each of the samples were stirred using a new stir stick. Any initial reactions were noted for the presence of carbonates. Third, the vials were placed in the hot water bath for 10 minutes, stirring every few minutes. Fourth, the samples were centrifuged and the excess HCL was decanted into an appropriate waste container. Finally, the samples were rinsed, centrifuged, and decanted twice using distilled water to remove the excess HCL.

The final stage of the pollen concentration process was a second 10 µm sieving of the samples to remove the finer particles within the samples that had been produced by the acid digestion processes. If the samples had not been adequately concentrated for pollen, then a second round of chemical processing and sieving was done to further concentrate the pollen. Depending on the amount of non-pollen residual material left in the sample after the second 10 µm sieving, the samples needed to undergo both chemical and sieving concentrations for a second time.

Pollen Grain Extraction

Once the final sieving had been completed, the samples were centrifuged and then picked for bisaccate pollen as described by (Mensing and Southon, 1999). A mouth operated pipetting system

was utilized to suck up and gather the conifer pollen from the samples. Processed samples were placed in petri dishes and a dissection microscope set at a 35X – 50X was used in order to differentiate between the pollen grains and the surrounding residual material. While extracting the pollen from the samples, distilled water was used to ensure the purity of the sample. Pollen was stored in 15 ml vials and shipped to a ^{14}C dating lab.